Green synthesis of ZrO$_2$ nanoparticles and nanocomposites for biomedical and environmental applications: a review

Thuan Van Tran$^{1}$ · Duyen Thi Cam Nguyen$^{1,2}$ · Ponnusamy Senthil Kumar$^3$ · Azam Taufik Mohd Din$^4$ · Aishah Abdul Jalil$^{2,5}$ · Dai-Viet N. Vo$^{1,4}$

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Abstract
Pollution and diseases such as the coronavirus pandemic (COVID-19) are major issues that may be solved partly by nanotechnology. Here we review the synthesis of ZrO$_2$ nanoparticles and their nanocomposites using compounds from bacteria, fungi, microalgae, and plants. For instance, bacteria, microalgae, and fungi secret bioactive metabolites such as fucoidans, digestive enzymes, and proteins, while plant tissues are rich in reducing sugars, polyphenols, flavonoids, saponins, and amino acids. These compounds allow reducing, capping, chelating, and stabilizing during the transformation of Zr$^{4+}$ into ZrO$_2$ nanoparticles. Green ZrO$_2$ nanoparticles display unique properties such as a nanoscale size of 5–50 nm, diverse morphologies, e.g. nanospheres, nanorods and nanochains, and wide bandgap energy of 3.7–5.5 eV. Their high stability and biocompatibility are suitable biomedical and environmental applications, such as pathogen and cancer inactivation, and pollutant removal. Emerging applications of green ZrO$_2$-based nanocomposites include water treatment, catalytic reduction, nanoelectronic devices, and anti-biofilms.

Keywords ZrO$_2$ nanoparticles · Green synthesis · Biomedical applications · Environmental remediation · ZrO$_2$-based nanocomposites

Introduction
Environmental pollution and climate change are presenting as two of the most globally critical issues in the context of the coronavirus pandemic (COVID-19) and circular economy (Muhammad et al. 2020; Ufnalska and Lichtfouse 2021). The advent of nanoscience and nanotechnology can solve these problems; thereby, holding the key to economic recovery, and environmental mitigation (Weiss et al. 2020; Tang et al. 2021). Nanomaterials play a central role in nanotechnology, raising the demands for sustainable development (Srivastava et al. 2021). The chemical synthesis of nanomaterials chiefly does not satisfy the strict requirements for this trend, and also mismatches with the principles of green chemistry. Currently, the green approaches for the synthesis of nanomaterials are of remarkable significance due to their eco-friendliness as well as cost-effectiveness (Tran et al. 2020a). This also pays the way for multiple applications of nanomaterials in the environmental mitigation, biomedical, and catalysis fields.

Zirconium (Zr) is classified as a transition metal element (d-block) in the titanium family (group IV) with the
atomic number 40. Zr does not basically capture neutrons, offering a potential metallic cladding for the fuel rods in the nuclear reactors (Cazado et al. 2021). Zirconium dioxide (ZrO₂) or zirconia is one of the highly stable oxides, created by thermalizing zirconium compounds (Hassan and Jalil 2022). Depending on the various synthesis routes, ZrO₂ can present in the crystalline phases involving monoclinic, tetragonal, and cubic (Zhang et al. 2018). Bulk ZrO₂ has a wide bandgap energy, typically ranging from 5.0 to 7.0 eV. Because of the ultrahigh stability and very low toxicity, ZrO₂ has exhibited an intensive range of practical technologies for heat-resistant ceramic superalloys (Wang et al. 2021), dental restorations (Chen et al. 2021), fuel cells (Rambabu et al. 2020), and heterogeneous catalysis (Jiang et al. 2020). Such promising utilizations make ZrO₂ an ideal nanomaterial, promoting the green strategy for synthesizing ZrO₂ nanoparticles.

In general, there are two major approaches to synthesize the ZrO₂ nanoparticles, involving top-down and bottom-up (Jadoun et al. 2021). The former implies the conversion of bulk material into thinner crystallites by the physical route. This means that it needs the participation of enormous mechanical energy sources such as milling, and ionic sputtering (Shrimal et al. 2020). As a result, top-down strategy brings many inevitable drawbacks of causing secondary impressions or intermediates, altering the physicochemical property and surface chemistry of as-synthesized nanoparticles (Indiarto et al. 2021). More importantly, nano-sized particles are mostly unattainable by top-down approach. Meanwhile, the latter implies the formation of particles by creating building blocks from ultra-small particles such as atoms or molecules, and then assembling them together. By this way, the nanostructured particles can be intentionally attainable under the control of fabrication conditions (Rana et al. 2020). The bottom-up strategy involves the physical (e.g., vapor decomposition, plasma irradiation, and ultrasonication), chemical (e.g., sol–gel, co-precipitation, chemical reduction, and hydrothermal), and biological (e.g., plant, fungi, algae, and bacteria) methods. However, the physical bottom-up method requires a high investment cost for operating instruments and consumes a large amount of heating or electric energies (Bolade et al. 2020). The chemical bottom-up method necessarily adopts toxic chemicals for its protocols, resulting in adverse impacts on the environment. Therefore, it may be greatly difficult for physical and chemical methods to attain a critical green strategy.

The biological bottom-up method is the most adequate for the green synthesis of ZrO₂ nanoparticles because it offers an effective, tunable, and eco-friendly approach (Shafey 2020). The biogenic synthesis uses low-cost and locally available sources such as plants or other biocompatible sources such as fungi, algae, and bacteria for ZrO₂ fabrication (Rana et al. 2020; Nguyen et al. 2021d). The biomolecules extracted from the biological sources play a vital role as extremely efficient bioreducing, biocapping, and biostabilizing agents, bringing excellent ZrO₂ production yields (Bandeira et al. 2020). Such biological substrates can safely replace almost highly expensive and toxic chemicals or energy-consuming physical instruments (Agarwal et al. 2017). Biological method for synthesis of ZrO₂ nanoparticles also match well with principle of sustainable and green chemistry (Yadi et al. 2018). Because of diminishing the potential risks from chemical and physical methods, generating no hazardous intermediates, and secondary pollutants, biogenic synthesis of ZrO₂ nanoparticles can be therefore considered as the green synthesis (Jadoun et al. 2021).

To our knowledge, there is only a limited number of relevant works (about twenty research articles) reported the green synthesis of ZrO₂ nanoparticles using various biological sources such as bacteria, fungi, and plant extract. Moreover, there is currently no comprehensive review on the systematic evaluations from the green synthesis to the applications of ZrO₂ nanoparticles and their nanocomposites. Therefore, the main objectives of this study are to discuss the current findings on green ZrO₂ preparation techniques and highlight their performance in the biomedical field and environmental remediation. Specifically, this review has the following organization: (i) green synthesis methods for ZrO₂ nanoparticles; (ii) structural characterizations of green ZrO₂ nanoparticles; (iii) biomedical, adsorption, catalytic, and other applications of green ZrO₂ nanoparticles; and (iv) emerging applications of green ZrO₂-based nanocomposites (Fig. 1). We briefly elucidate some challenging and prospective aspects of green ZrO₂ nanocomposites for expanding their potential applications.

**Green synthesis of ZrO₂ nanoparticles**

**Bacteria**

Bacteria are among the fastest-growing microorganisms under mild cultivation conditions in temperature, pressure, and pH (Ram et al. 2020). They are therefore the favorable biofactories for the synthesis of ZrO₂ nanoparticles. During the metabolism activities, bacteria secrete some enzymes and proteins extracellularly or intracellularly, which possibly aid the bioreduction, biocapping, and biostabilization of zirconium ions (Farig et al. 2017). Underlining mechanism of forming ZrO₂ nanoparticles by bacterial communities endures further complex steps including (i) biosorption and (ii) bioreduction (Saravanan et al. 2021). The former step initiates with trapping zirconium ions on the bacterial cell surface through the physical and chemical interactions (e.g., electrostatic, hydrogen bonding, ionic exchanging, and chelating) (Gahlawat and Choudhury 2019). Specifically,
the secretion of macromolecules such as proteins, polysaccharides containing many negatively charged functional groups enriches the surface chemistry of bacterial cell wall (Patil and Kim 2018). This creates the attraction of Zr$^{4+}$ ions towards bacterial cell surface. The latter step takes main responsibility for reducing Zr$^{4+}$ ions into ZrO$_2$ nanoparticles. Bioactive compounds such as acid amines, proteins, etc. in the extracellular or intracellular environment can participate in the formation and stabilization of ZrO$_2$ nanoparticles (Narayanan and Sakthivel 2010).

Suriyaraj et al. (2019) reported the one-step biofabrication of ZrO$_2$ nanoparticles using extremophilic Acinetobacter sp. bacterial community. This green source was firstly cultivated in glucose solution, and added by 100 mM ZrOCl$_2$.8H$_2$O for incubation for 12 h. Interestingly, highly crystalline ZrO$_2$ nanomaterial was facilely precipitated under room-temperature condition (37 °C). Moreover, this

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**Fig. 1**  Green synthesis of ZrO$_2$ nanoparticles and their applications in biomedical, adsorption, catalysis, and nanocomposite fabrication. The plant tissues including flowers, fruits, seeds, leaves, roots, etc. possess many phytochemicals such as polyphenols, saponins, quercetin, and gallic acid. Microorganisms including bacteria, fungi, algae, etc. can secrete biomolecules, metabolites, enzymes, and proteins. These phytochemicals and biomolecules participate in the green synthesis of ZrO$_2$. 

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study found biocompatibility and ineligible cytotoxicity with mouse fibroblast cells. The fluorescein staining was carried out to confirm the major role of extracellular proteins released from *Acinetobacter* sp. for biosynthesis of ZrO$_2$. Very recently, Ahmed et al. (2021) successfully synthesized zirconia nanoparticles using *Enterobacter* sp. strain. Notably, the bacterial strain was facilely isolated from the paddy soils, and then, incubated at room temperature. After the bacterial purification, 5 mM ZrOCl$_2$·8H$_2$O was added to the supernatant under agitating incubation for 24 h. Visible precipitation was finally formed at the bottom of the flask. It was proposed that nitrate reductase enzyme was in charge of reducing Zr$^{4+}$ ions into zirconia and protein secreted by *Enterobacter* sp. acts as a capping agent (Fig. 2). As biosynthesized ZrO$_2$ nanoparticles revealed the promising antifungal activity with 75.5% inhibition against Pestalotiopsis Versicolor disease pathogen. These publications have demonstrated the great potentials and biocompatibility of the bacterially synthesized ZrO$_2$ nanoparticles.

**Fungi**

Fungi include the microbial organisms that absorb nutrients by decomposing organic matters, without the photosynthesis pathways (Chu et al. 2021). They play the principal role in nutrient cycling and exchanging as well as mitigation of metals-contaminated soils (Riaz et al. 2021). During these processes, fungal communities secrete digestive enzymes into their foods to acquire the nutrients essential for growth (Ferreira et al. 2020). Taking advantage of such features, fungi can be ideal biotemplates for the synthesis of ZrO$_2$ nanoparticles. Basically, the mechanism for green synthesis of ZrO$_2$ nanoparticles using fungal species is closely similar to that using bacterial species. However, the utilization of fungi for ZrO$_2$ production offers many advantageous points (Narayanan and Sakthivel 2010). Firstly, they can endure the harsh synthesis conditions such as flow pressure or agitation in the bioreactor, easily handling the fabrication of ZrO$_2$ (Abinaya et al. 2021). Secondly, fungi have accelerated growth in controllable ways, i.e., through adjusting their nutrient ingredients. Ultimately, almost all fungi are better resistant to genetic or environmental mutations, facilitating the culturing process (Bansal et al. 2011). These features of extracellular environment and aid the biocapping and biostabilization of zirconia nanoparticles. Reprinted with the permission of Elsevier from reference (Ahmed et al. 2021). Abbreviations: ZrONPs, zirconia nanoparticles; NAD$^+$, an oxidized form of nicotinamide adenine dinucleotide; NADH, a reduced form of nicotinamide adenine dinucleotide.
fungal species are more favorable for biosynthesis of ZrO2 than other biological synthesis methods such as bacteria and plants.

Ghomi et al. (2019) reported that the formation of ZrO2 by extracellular secretion of Penicillium fungal species including P. aculeatum, P. notatum, and P. purpurogennon. Under the optimum conditions at pH 9, and 1.5 mM Zr4+, the solution color changed from pale yellow into deep yellow after seven-day incubation, affirming the generation of colloidal ZrO2. In terms of the ZrO2 biofabrication mechanism, the enzymes present in the fungal supernatants may aid to transform Zr4+ ions into ZrO2 nanoparticles. Bansal et al. (2004) cultured another fungal species named Fusarium oxysporum for the successful biosynthesis of ZrO2 nanoparticles from the ZrF6 2− source at room temperature. To gain insight into the major role of proteins secreted from this fungus for the extracellular hydrolysis of ZrF6 2−, the resulting filtrates were lyophilized to test for hydrolytic activity. The findings confirmed the cationic nature of proteins to bind easily to ZrF6 2− anions, which could not be found in other fungal species (e.g., C. lunata, C. gloeosporioides, Phomopsis sp., A. niger). More importantly, ZrF6 2− anions showed a non-toxicity to Fusarium oxysporum, opening the enormous prospects for large-scale biosynthesis of ZrO2 nanoparticles. To sum up, the highlighted eco-friendly, biocompatible, and energy-conserving advantages of the fungal biosynthesis of ZrO2 nanoparticles are superior to chemical methods, and hence, should not be overlooked.

Algae

Great difference from fungal species, algae are a group of dominantly aquatic, eukaryotic, and autotrophic organisms that acquires their nutrients from the photosynthetic processes. Algae use chlorophyll for their primary photosynthesis; therefore, they are sometimes considered as terrestrial plants. Some algal species such as brown algae, or seaweeds excrete fucoidans from their cell walls (Filote et al. 2021). Fucoidans refer to multifunctional polysaccharides containing a substantial amount of fucose and sulfated esters, exhibiting many bioactivities involving antiviral, antioxidant, and anticoagulant (Ponce and Stortz 2020). These biomolecules can therefore act as ideal bioreducers for the green synthesis of ZrO2. Indeed, Kumaresan et al. (2018) successfully bio-prepared tetragonally nanostructured ZrO2 from the aqueous extract of seaweed Sargassum wightii. The biosynthesis procedure was tunable with the grind of ZrO(NO3)2·H2O with Sargassum wightii extract within 20 min. ZrO2 nanoparticles could be formed after the calcination at 400 °C. This nanomaterial showed the spherical morphology with the particle size of 5 nm and was utilized for investigating several potential antibacterial activities against gram-positive and gram negative-bacteria. The researchers demonstrated that large surface area and nanosize (4.8 nm) of ZrO2 obtained from this green strategy contributed considerably to enhancing growth inhibitory effects against antibacterial pathogens. Although many works reported the use of algae for biosynthesizing various nanoparticles, there is still an exception for ZrO2. It is also necessary to elucidate the underlying mechanisms for the formation of ZrO2 nanoparticles using algal species.

Plants

Plants are the most prevalent, locally available, highly cost-effective resource for biosynthesis of ZrO2 nanoparticles (Yadi et al. 2018). Compared with other green materials such as bacteria, fungi, and algae, the use of plants as biotemplates attains many notable benefits (Saravanan et al. 2021). Firstly, there are less risks to health than microbial ZrO2 synthesis strategies. Almost plants are substantially more benign than bacteria or fungi. Some microbial species can even secrete toxins or metabolites during cultivation (Yang and Chiu 2017; Xu et al. 2020). Secondly, the botanical synthesis of ZrO2 nanoparticles is considerably accelerated in comparison with microbial routes. The bioreduction of Zr4+ by biomolecules from plant extract occurs within a few minutes to several hours, while microbial communities mostly prolong these bioprocesses by many days under ambient incubation (Vijayaraghavan and Ashokkumar 2017). This results in the higher kinetic of overall ZrO2 production process. Thirdly, the synthesis of ZrO2 nanomaterial by plant extracts is controllable and easy to manipulate. Diverse parts of plants such as leaves, barks, roots, flowers, etc. can be effortlessly collected for biosynthesis of ZrO2 nanoparticles. The green solvents for extracting phytochemical compounds from plant tissues are mainly water and ethanol. Meanwhile, the microbial culture needs to be carried out under strict conditions to avoid the generation of colonies, unfavorable to ZrO2 fabrication.

Phytochemicals involving reducing sugars, polyphenols, flavonoids, saponins, and amino acids are inherent in the plant tissues such as flowers, leaves, and roots (Fig. 3a). These compounds are known as strong antioxidants or bioreductants, which can play pivotal roles as biocapping, biochelating, bioreducing, and biostabilizing agents for the synthesis of ZrO2 nanoparticles (Fig. 3b). The phytochemicals participate in the formation and stabilization of octahedral complex of Zr2+-phytochemicals. The presence of phytochemicals also aids to hamper the aggregation of ZrO2 nanoparticles during thermal calcination. This may boost the surface area of ZrO2 nanoparticles, which are conducive to their catalytic and adsorption properties. Consequently, the phytochemicals in plant extract can exhibit a range of important functionalities including replacing hazardous,
Fig. 3 Phytochemicals in plant extracts (a), and proposed mechanism for the biosynthesis of ZrO$_2$ nanoparticles using these phytochemicals (b). Phytochemicals act as bioreductants to convert Zr$^{4+}$ into octahedral complex [Zr(H$_2$O)$_6$]$^{2+}$. This enhances the stability and hinders the clustering of zirconium complexation during chelation with phytochemicals Zr$^{4+}$–phytochemicals. The calcination of Zr$^{4+}$–phytochemicals complex produces ZrO$_2$ nanoparticles in tandem with the release of CO$_2$, H$_2$O, N$_2$, and many decomposed products.

Table 1 Synthesis of green ZrO$_2$ nanoparticles using plants

| Plant extract and zirconium inputs | Complexation step | Calcination step | Refs. |
|-----------------------------------|------------------|-----------------|-------|
|                                   | Zr$^{4+}$/extract (v/v) | Temperature | Time | Temperature | Time |                                   |
| Euclea natalensis                 | Roots             | ZrOCl$_2$      | 1:1  | Not reported | 3 h  | 550 °C | 3 h |
| Aloe vera                         | Leaves            | ZrOCl$_2$      | Not reported | Room | 4 h  | 500 °C | Not reported |
| Salvia rosmarinus                 | Leaves            | ZrOCl$_2$      | 1:4  | 70 °C        | 1 h  | 600 °C | 2 h |
| Sapindus mukorossii               | Pericarp          | ZrOCl$_2$      | 1:1  | 60 °C        | 3–4 h| 500–700 °C | Not reported |
| Wrightia tinctoria                | Leaves            | ZrOCl$_2$      | 1:1  | 75 °C        | 3–4 h| 800 °C | Not reported |
| Ficus benghalensis                | Leaves            | ZrOCl$_2$      | 1:1  | Microwave    | 15 min| 500 °C | 3 h |
| Moringa oleifera                 | Leaves            | ZrOCl$_2$      | 1:1  | 60 °C        | 3–4 h| 700–800 °C | Not reported |
| Heliantus annuus                  | Seeds             | ZrOCl$_2$      | 1:5  | Not reported | 2–3 h| 600 °C | 4 h |
| Tinospora cordifolia              | Leaves            | ZrOCl$_2$      | 10:1 | 55 °C        | 40 min| Not reported | Not reported |
| Nepheleium lappaceum              | Fruit             | Zr(NO$_3$)$_2$ | 7:1  | 180 °C       | 18 h | 500 °C | 4 h |
| Nyctanthes arbor-tristis          | Flower            | ZrOCl$_2$      | 2:1  | Not reported | 2 h  | 300–500 °C | 3 h |
expensive chemical antioxidants or reductants, and shortening the number of complicated synthesis steps.

The general botanical biosynthesis of ZrO₂ nanoparticles initiates with the collection and pretreatment of plant tissues to eliminate the irrelevant parts (Table 1). The procedure is followed by extracting phytochemicals using several common kinds of solvents such as water, ethanol, or hexane from cell walls (Saraswathi and Santhakumar 2017; Goyal et al. 2021; Alagarsamy et al. 2022). Water is a prevalently used, and highly polar extractor which recovers the aqueously soluble phytochemical compounds such as gallic acid, polyphenols, glucose, etc. while ethanol exhibits a poorer polarity than water, but higher than hexane. There are two major steps including complexation and calcination to reach the final ZrO₂ products. The former is carried out under the incorporation of plant extract into zirconium source, mainly ZrOCl₂·8H₂O. The proper temperature and time the incorporation of plant extract into zirconium source, into ZrO₂ nanoparticles.

The bandgap energy of a semiconductor reflects the energy disparity between the top of valence band and the bottom of conduction band, or the minimum energy to excite an electron from the valence band to the conduction band (Makuta et al. 2018). It can be determined from diffuse reflectance spectra. While the bulk ZrO₂ nanoparticles have a wide bandgap energy (~5.0 eV), green ZrO₂ nanoparticles exhibit values ranging from 3.7 to 5.5 eV (Table 2). Indeed, Goyal et al. (2021) fabricated ZrO₂ nanoparticles using methanolic extract of Helianthus annuus seeds with very narrow bandgap (3.7 eV). Al-Zaqri et al. (2021) found the same small value of ZrO₂ nanoparticles biosynthesized from Wrightia tinctoria leaf extract. They also gave an assumption about more contribution from external surface defects ZrO₂ nanoparticles. Meanwhile, obtaining higher bandgap energies (5.4–5.5 eV) of other green ZrO₂ nanoparticles may be ascribed by quantum confinement effects (Gowri et al. 2014; Alagarsamy et al. 2022). The bandgap augments for the smaller size nanoparticles due to the deformity phenomena. Overall, biologically synthesized ZrO₂ nanoparticles acquire sufficient gap energies, capable of exhibiting their excellent catalytic and biomedical activities through the formation of reactive oxygen species.

Properties of green ZrO₂ nanoparticles

Crystallinity

Bulk ZrO₂ nanoparticles exist in three common crystalline phases involving monoclinic, tetragonal, and cubic. Biologically synthesized ZrO₂ nanoparticles can also attain such crystalline phases based on X-Ray diffraction analysis (Table 2). Specifically, Davar et al. (2018) reported a cubic phase of ZrO₂ nanoparticles bioproduced from Salvia Ros-marinus plant extract with the reflecting planes at (111), (200), (220), (311), (222), and (400). Another study indicated the monoclinic phase of ZrO₂ synthesized from Helianthus annuus plant extract (Goyal et al. 2021). However, tetragonal phase is the most prevalent single-crystal phase of green ZrO₂ nanoparticles. Green ZrO₂ nanoparticles can also have multiphase crystallinity, which includes at least two phases (Sathishkumar et al. 2013; Debnath et al. 2020; Isac-franklin et al. 2020). The presence of crystal phase in green ZrO₂ possibly affects their electronic properties, optical band gap, and surface energy, and stability. It is therefore essential for further studies to clarify the role of synthesis conditions, e.g., the ratio between zirconium and plant extract in the formation of ZrO₂ phase.

Optical bandgap

Many physicochemical techniques such as X-Ray diffraction, dynamic light scattering, transmission electron microscopy, etc. can be used to measure the average particle size of ZrO₂ nanoparticles. From the database of previous studies (Table 2), the particle size of ZrO₂ nanoparticles is distributed from 5 to 150 nm, commonly less than 50 nm. Compared with chemical approaches, biological methods generally create ZrO₂ nanoparticles with considerably smaller sizes (Suriyaraj et al. 2019; Ahmed et al. 2021). It can be understandable that biomolecules from the secretion of microbial sources, or phytochemicals from plant extracts lessen the assembly of ZrO₂ nanoparticles (Kumaresan et al. 2018; Silva et al. 2019; Debnath et al. 2020). Small-size ZrO₂ nanoparticles offer many biomedical benefits since they facilitate the penetration into cell walls, performing better antibacterial, antifungal, and anticancer activities (Khatoon et al. 2015).

Morphology

The shape of ZrO₂ nanocrystals biosynthesized from biological routes can be explored by scanning/transmission electron microscopy analysis. Accordingly, green ZrO₂ nanoparticles can present a wide range of morphological properties such as nanospheres (mainly), nanochains,
nanorods, semi-nanospheres, nano-sized ovals, and nano-flakes (Fig. 4). Generally, the ZrO$_2$ morphology is significantly dependent on the synthesis conditions including the ratio between zirconium source and green extract. Spherical ZrO$_2$ can be synthesized by using microbial sources with the long incubation (1–3 days) at room temperature or by several botanical sources such as Euclea natalensis roots, Aloe vera leaves, Ficus benghalensis leaves, Moringa oleifera leaves,

| Green material | Species                  | Particle size (nm) | Morphology      | Crystal phase     | Optical band gap (eV) | Surface chemistry                                      | Refs.                  |
|----------------|--------------------------|--------------------|-----------------|-------------------|-----------------------|--------------------------------------------------------|------------------------|
| Alga           | Sargassum wightii        | 5.0                | Nanosphere      | Tetragonal        | 4.4                   | Hydroxyl, carboxylate                                   | Kumaresan et al. (2018) |
| Bacterium      | Enterobacter sp.         | 33–75              | Nanosphere      | Not reported      | Not reported          | Hydroxyl, carboxyl, alkene                             | Ahmed et al. (2021)    |
| Bacterium      | Acinetobacter sp.        | 44±7               | Nanosphere      | Monoclinic, tetragonal | 4.9             | Hydroxyl                                               | Suriyaraj et al. (2019) |
| Bacterium      | Pseudomonas aeruginosa   | 6.14–15            | Nanosphere      | Monoclinic, tetragonal | Not reported       | Hydroxyl, zirconium hydroxide                          | Debath et al. (2020)   |
| Fungus         | Penicillium              | < 100              | Nanosphere      | Not reported      | Not reported          | Hydroxyl, carbomyl, amine                              | Ghomi et al. (2019)    |
| Fungus         | Fusarium solani          | Not reported       | Nanosphere      | Tetragonal        | Not reported          | Hydroxyl, carbomyl                                     | Kavitha et al. (2020)  |
| Plant          | Euclea natalensis        | 5.9–8.54           | Nanosphere      | Monoclinic, tetragonal | Not reported | Hydroxyl                                               | Silva et al. (2019)    |
| Plant          | Aloe vera                | < 50               | Nanosphere      | Tetragonal        | 5.42                  | Hydroxyl, carbomyl, zirconium hydroxide                | Gowri et al. (2014)    |
| Plant          | Carcuma longa            | 41–45              | Nanochain       | Monoclinic, rhombohedral | Not reported | zirconium hydroxide                                    | Sathishkumar et al. (2013) |
| Plant          | Aloe vera                | 18–42              | Not reported    | Face centered cubic | Not reported | Hydroxyl, carbomyl, amine, alkene                       | Prasad et al. (2014)   |
| Plant          | Rosmarinus officinalis   | 12–17              | Semi-nanosphere | Cubic             | Not reported          | Hydroxyl, carboxylic, alkyl                             | Davar et al. (2018)    |
| Plant          | Sapindus mukorossi       | 5–10               | Nanosphere      | Tetragonal        | 5.535                 | Hydroxyl                                               | Alagarsamy et al. (2022) |
| Plant          | Wrightia tinctoria       | 17                 | Nanosphere      | Monoclinic, tetragonal | 3.78            | Hydroxyl, alkyl, carboxylic acid                       | Al-Zaqri et al. (2021) |
| Plant          | Lagerstroemia speciosa   | 56.8               | Oval            | Tetragonal        | Not reported          | Hydroxyl, carbomyl, amine                              | Saraswathi and Santhakumar (2017) |
| Plant          | Ficus benghalensis       | 14.7               | Nanosphere      | Monoclinic, tetragonal | 4.9              | Hydroxyl, alkyl, carbomyl                              | Shinde et al. (2018)   |
| Plant          | Moringa oleifera         | < 10               | Nanosphere      | Not reported      | Not reported          | Hydroxyl                                               | Annu et al. (2020)     |
| Plant          | Helianthus annuus        | 35.45              | Nanosphere      | Monoclinic        | 3.7                   | Carboxyl, alkyl, amine                                 | Goyal et al. (2021)    |
| Plant          | Laurus nobilis           | 20–100             | Nanosphere      | Monoclinic, tetragonal | Not reported | Hydroxyl                                               | Chau et al. (2021)     |
| Plant          | Tinospora cordifolia     | 73                 | Nanosphere      | Not reported      | Not reported          | Hydroxyl, carbomyl                                     | Joshi et al. (2021)    |
| Plant          | Nephelium laplaceum      | 50                 | Nanorod         | Monoclinic, cubic | Not reported          | Hydroxyl                                               | Isacfranklin et al. (2020) |
| Plant          | Nyctanthes arbor-tristis | < 150              | Nano-flake      | Tetragonal        | 5.31                  | Not reported                                           | Gowri et al. (2015)    |
Helianthus annuus seeds, Tinospora cordifolia leaves, and Laurus nobilis leaves (Table 2). Meanwhile, other ZrO₂ morphologies are relatively rare, mostly synthesized by plant sources. For example, Sathishkumar et al. (2013) obtained ZrO₂ nano-chains by the hydrolysis of ZrF₆²⁻ in the presence of Curcuma longa tuber extract. Gowri et al. (2015) also successfully biosynthesized ZrO₂ nanoflakes by the hydrolysis of ZrOCl₂·8H₂O using aqueously soluble carbohydrates extracted from Nyctanthes arbor-tristis flower. Semi-spherical and oval ZrO₂ can be formed from many plant extracts such as Salvia Rosmarinus and Lagerstroemia speciose, respectively (Saraswathi and Santhakumar 2017; Davar et al. 2018). More importantly, diverse morphologies of ZrO₂ morphologies can bring many benefits for practical applications. As an example, (Isacfranklin et al. 2020) indicated that ZrO₂ nanorods possess sufficient cellular uptake and toxicity, offering the prospect for biomedical application.

**Surface chemistry**

The surface functionalization by chemical groups aids to extend the applications of nanomaterials (Tran et al. 2021). Surface chemistry of ZrO₂ nanoparticles can be identified by several physicochemical techniques such as Fourier-transform infrared spectroscopy and X-ray photoelectron spectroscopy or quantified by Boehm titrations (Nguyen et al. 2021b). The surface functional groups on ZrO₂ nanoparticles may be originated from the phytochemical compounds. Considering the calcination of the complex of Zr²⁺–phytochemicals, almost all organic components are decomposed into volatiles or simple products such as CO₂, N₂, and H₂O. A tiny proportion of phytochemicals may still remain due to the chelating with zirconium ions. They can be thermally modified to generate simpler biomolecules with more diverse functional groups such as carbonyl, amine, carboxylate, and alkyl (Table 2). However, hydroxyl groups are detected partly owing to H₂O-adsorbed surface of ZrO₂ (Kumaresan et al. 2018; Silva et al. 2019; Annu et al. 2020). The functionalization of ZrO₂ surface possibly can lead to the improvement of adsorption efficiency towards many pollutants through many key interactions such as electrostatic attraction, hydrogen bond, Yoshida hydrogen bond, and π–π interaction (Suresh et al. 2015; Nguyen et al. 2021c).
Surface area

Theoretically, surface area of nanomaterials links closely to the number of active sites on the edges, resulting in a significant influence on their catalytic activity (Jiang et al. 2011). The higher surface area of ZrO2 nanoparticles can also facilitate the better favorable adsorption process. The bio compounds such as proteins, enzymes, polysaccharides, polyphenols, etc. present in the biological source extract chelating with zirconium ions can prevent the aggregation of ZrO2 nanoparticles during the thermolysis. This enhances the surface area and the number of active sites of ZrO2 nanoparticles. For example, Shinde et al. (2018) reported the surface area (88 m²/g) of ZrO2 nanoparticles biosynthesized from Ficus benghalensis leaf extract using BET (Brunauer, Emmett and Teller) analysis. It was also witnessed an enhancement of the photocatalytic activities of ZrO2 to methylene blue (91%) and methyl orange (69%) for 240 min. These results of green ZrO2 were remarkably higher than those obtained by the chemical methods such as anodization in H₂O₂/NH₄F/ethylene glycol electrolyte (Rozana et al. 2017), decomposition of Zr(OH)₄-urea complex (Sudrajat et al. 2016), polymer-assisted sol–gel synthesis (Dhandaj et al. 2016).

Stability

Compared with chemically produced nanomaterials, biologically produced ZrO2 nanoparticles not only exhibit good dispersion but also show better stability. For example, Sathishkumar et al. (2013) found the surprising stability of zirconia nanoparticles up to five months after green fabrication. This phenomenon may be elucidated by the presence of phytochemicals acting as efficient biostabilizers and biocapping agents. The stability of ZrO2 nanoparticles can be assessed by the zeta potential measurements. Basically, the zeta potential values are different with zero, normally minimum ± 30 mV, indicating the high stability of ZrO2 nanoparticles (Jameel et al. 2020). Indeed, Suriyaraj et al. (2019) demonstrated the well-dispersed, anti-clustering, and nanostructured ZrO2 suspension by the measurement of zeta potential (36.5 ± 5.5 mV). Chau et al. (2021) also measured the high zeta potential (−32.8 mV) of ZrO2 nanoparticles biosynthesized from Laurus nobilis leaf extract by dynamic light scattering analysis, proposing the good anti-sedimentation of the nanomaterials for antimicrobial activities.

Applications of green ZrO₂ nanoparticles

Biomedical applications

Biomedical compatibility of green ZrO₂ nanoparticles

Bacterial and fungal species attack the immune system of humans and animals and become the major factors causing infections and even deaths (Humbal et al. 2018). Many antibiotics and antifungal pharmaceuticals have been developed over the centuries, but mankind is increasingly encountering the huge problems of antibiotic resistance (Kovalakova et al. 2020). Among the efforts to search for alternatives to antibiotics, bionanotechnology exhibits its high reliability and performance contributing proactively to the abatement of infections (Ong et al. 2018). Many metallic nanomaterials (e.g., silver nanoparticles) possess their inherent antibacterial and antifungal activities; hence, they are widely utilized in biomedical fields (Abbasi et al. 2014). However, the lack of biocompatibility, biostability, and effectiveness may restrain the biomedical application of these nanomaterials. Green ZrO₂ nanoparticles biosynthesized from environmentally friendly botanical and microbial sources show unique properties, adequate for fabricating biomedical devices. Based on the published literature, this section will discuss some potential activities of green ZrO₂ nanoparticles.

Antibacterial activity

The plasma membrane of bacteria is mostly constituted of proteins (e.g., peptidoglycan macromolecules) that charge negatively (Li et al. 2019). ZrO₂ nanoparticles charge positively on their surface, leading to electrostatic interactions with bacterial membranes (Xing et al. 2018). This facilitates the biosorption and bioaccumulation of ZrO₂ nanoparticles on the cell walls. With the nanoscale size, high surface area, and good biocompatibility of green ZrO₂ nanoparticles, they easily pass through cell membrane, inhibit the key metabolic functions, and finally deactivate bacterial cells (Kumaresan et al. 2018). It is suggested that the generation of reactive oxygen species (e.g., ·O₂⁻, and ·OH) is the underlying mechanism (Akintelu and Foloruso 2020). Accordingly, these species damage the genetic material such as deoxyribonucleic and ribonucleic acids, disordering the transcription and translation processes in bacteria (Slavin et al. 2017). Bacterial death occurs as a result of cell division failure.

There are many publications that reported the excellent antibacterial activity of green ZrO₂ nanoparticles against both gram-negative and positive bacteria (Table 3). For example, Gowri et al. (2015) investigated the antibacterial activity against E. coli and S. aureus using ZrO₂ biosynthesized from Nyctanthes arbor-tristis plant extract. The
| Green material | Species            | Type of activity | Target                                      | Main findings                                                                                     | Ref                      |
|----------------|--------------------|------------------|---------------------------------------------|---------------------------------------------------------------------------------------------------|--------------------------|
| Plant          | *Nyctanthes arbor-tristis* | Antibacterial    | Gram negative (*E. coli*), Gram positive (*S. aureus*) | Zones of inhibition against *E. coli* (17 mm), *S. aureus* (15 mm) for ZrO₂ NPs, and those against *E. coli* (30 mm), and *S. aureus* (22 mm) for ZrO₂-treated cotton fabric | Gowri et al. (2015)     |
| Fungus         | *Penicillium*      | Antibacterial    | Gram negative (*E. coli*, *P. aeruginosa*) and Gram positive (*S. aureus*) | Minimum inhibitory concentrations for *E. coli* (0.75 mM), *P. aeruginosa* (0.375 mM), but not found for *S. aureus* | Ghomi et al. (2019)     |
| Plant          | *Wrightia tinctoria* | Antibacterial    | Gram negative (*E. coli*, *P. aeruginosa*) and Gram positive (*S. aureus*, *B. subtilis*) | Zones of inhibition observed against *E. coli* (22 ± 0.5 mm), *P. aeruginosa* (21 ± 0.3 mm), *S. aureus* (21 ± 0.4 mm), and *B. subtilis* (20 ± 0.2 mm) at a ZrO₂ concentration of 10 μg/mL. Strong antibacterial may be due to the flavonoids, protein, tannins, alkaloids, amino acids and carbohydrates present in the leaf extract | Al-Zagri et al. (2021)  |
| Plant          | *Moringa oleifera* | Antibacterial    | Gram negative (*E. coli*, *P. aeruginosa*) and Gram positive (*S. aureus*, *B. subtilis*) | Zone of inhibition against *E. coli* (4 mm), *P. aeruginosa* (10 mm), *S. aureus* (not observed), and *B. subtilis* (9 mm) at a concentration of 20 mg | Annu et al. (2020)       |
| Plant          | *Helianthus annuus* | Antibacterial    | Gram negative (*E. coli*, *P. aeruginosa*) and Gram positive (*S. aureus*, *K. pneumonia*) | Zone of inhibition against *E. coli* (13 mm), *P. aeruginosa* (13.5 mm), *S. aureus* (12.0 mm), and *K. pneumonia* (12.5 mm) at a ZrO₂ concentration of 100 μg/mL. | Goyal et al. (2021)      |
| Alga           | *Sargassum wightii* | Antibacterial    | Gram negative (*E. coli*, *S. typhi*) and Gram positive (*B. subtilis*) | Zone of inhibition (mm) against *E. coli* (19 mm), *S. typhi* (19 mm) and *B. subtilis* (21 mm) at a ZrO₂ concentration of 15 μg/mL. | Kumaresan et al. (2018) |
| Plant          | *Enterobacter sp.* | Antifungal       | Bayberry twig blight pathogen *P. versicolor* | Wide antifungal inhibition zone (25.18 ± 1.52 mm) against *P. versicolor* disease at 20 μg mL⁻¹ zirconia nanoparticles | Ahmed et al. (2021)     |
| Plant          | *Laurus nobilis*   | Antibacterial, antifungal | Gram negative (*K. pneumonia*, *E. coli*), Gram positive (*B. subtilis*, *S. aureus*), and fungi strain (*A. niger*) | Zone of inhibition (mm) against *B. subtilis* (14 mm), *S. aureus* (13 mm), *K. pneumonia* (15 mm), *E. coli* (14 mm), and *A. niger* (15 mm) | Chau et al. (2021)      |
| Plant          | *Tinospora cordifolia* | Antibacterial, antifungal | Gram negative (*P. aeruginosa*, *E. coli*, *A. fumigatus*), Gram positive (*B. subtilis*, *S. mutans*), and fungi strain (*A. niger*) | Zone of inhibition against *P. aeruginosa* (32 mm), *E. coli* (34 mm), *A. fumigatus* (34 mm), *B. subtilis* (36 mm), *S. mutans* (28 mm), and *A. niger* (32 mm) | Joshi et al. (2021)     |
inhibition zones were used to measure the antibacterial activity of green ZrO₂ nanoparticles. The values were significantly improved in the case of the cotton fabric treated with ZrO₂ nanoparticles. With very low green ZrO₂ concentration (10 μg/mL), Al-Zaqri et al. (2021) revealed good antibacterial activities against gram-negative and positive bacteria. They assumed the major role of biomolecules presented in Wrightia tinctoria leaf extract in improving the properties of green ZrO₂ nanoparticles, e.g., small size particles and high surfaces, enhancing their antibacterial activities. In addition to plant extract, ZrO₂ can be biosynthesized from other green materials such as fungi and algae for investigating antibacterial activities. Indeed, Kumaresan et al. (2018) reported the use of Sargassum wightii alga for the synthesis of ZrO₂, exhibiting the wide zones of inhibition (19–21 mm) against E. coli, S. typhi, and B. subtilis. Meanwhile, Ghomi et al. (2019) utilized the fungus Penicillium to synthesize the ZrO₂ nanoparticles. Minimum inhibitory concentrations against P. aeruginosa and E. coli were found, at 0.375, and 0.75 mmol/L, respectively.

Antifungal activity

Basically, the ZrO₂ nanoparticles may have the inhibition mechanism against fungi as same as that against bacteria (Ahmed et al. 2021). The main mechanism of the disruption of fungal cell division may rely on the generation of reactive oxygen species (e.g., ·O₂⁻, and ·OH) and free radicals (Akintelu and Folorunso 2020). With the high biocompatibility, green ZrO₂ nanoparticles can pervade through the fungal cell membranes by endocytosis. In the intracellular environment, ZrO₂ nanoparticles account for detrimental functions such as ribosomal, chromosomal, and mitochondrial damages through the formation of reactive oxygen species (Fig. 5). Moreover, ZrO₂ nanoparticles possibly break the integrity of fungal cell by changing the physicochemical conditions to cause cell stress and leaking the intracellular components (Reddy et al. 2015).

Although the mechanisms of damaging the fungal cells by ZrO₂ nanoparticles have not been fully understood, many attempts were still made to demonstrate their antifungal efficiency. Ahmed et al. (2021) obtained the promising practical outcomes of ZrO₂ nanoparticles biosynthesized from Enterobacter sp. against bayberry fungal pathogen. Moreover, the antifungal inhibition zones were very wide 25.18 ± 1.52 mm at the dilute concentration of ZrO₂ (only 20 μg/mL). They also observed the clear deconstruction of P. Versicolor fungal cell by extracellular leakage of deoxyribonucleic acid (DNA) and proteins under microscopy imaging techniques. Several works clarified the strength of green ZrO₂ for inhibiting both fungal and bacterial species (Chau et al. 2021; Joshi et al. 2021). Regardless of some limitations of the scope of fungal and bacterial species, those may
| Green material | Species | Type of treatment | Target pollutants | Main findings | Refs. |
|---------------|---------|------------------|-------------------|--------------|-------|
| Bacterium     | *Pseudomonas aeruginosa* | Adsorption | Tetracycline | Optimum pH 6.0 High adsorption capacity of 526.32 mg/g, adsorption model fitted best with Langmuir model Reusability up to 5 cycles (81.55% after 5th cycle) | Debnath et al. (2020) |
| Plant         | *Euclea natalensis* | Adsorption | Tetracycline | The best adsorptive capacity of 30.45 mg/g was obtained by response surface methodology | Silva et al. (2019) |
| Plant         | *Aloe vera* | Adsorption | Fluoride | Exothermic and spontaneous adsorption between 20 and 50 °C Nearly 99% F⁻ ions were adsorbed by ZrO₂-based adsorbent Chemisorption capacity of 96.58 mg/g | Prasad et al. (2014) |
| Plant         | *Sapindus mukorossi* | Adsorption | Methylene blue | Optimum adsorption conditions at pH 10, adsorbent dosage of 0.3 g, initial methylene blue concentration of 20 mg/L, and average time of 300 min 94% removal efficiency for methylene blue dye and adsorptive capacity of 23.26 mg/g Good recyclability: 0.1 M HCl as an efficient eluent, and three consecutive cycles | Alagarsamy et al. (2022) |
| Plant         | *Wrightia tinctoria* | Catalytic degradation | Reactive yellow 160 dye | 94.58% degradation for Reactive yellow 160 azo dye 0.9837 min⁻¹ for first order rate constant (k₁) | Al-Zaqri et al. (2021) |
| Plant         | *Lagerstroemia speciosa* | Catalytic degradation | Methyl orange | Degradation percentage was at 94.58% after irradiating under the sunlight for 290 min | Saraswathi and Santha-kumar (2017) |
| Plant         | *Ficus benghalensis* | Catalytic degradation | Methylene blue | Optimum conditions: catalyst loading of 1.5 g/L at pH 7 Removal of 91.22% after 240 min | Shinde et al. (2018) |
| Plant         | *Ficus benghalensis* | Catalytic degradation | Methyl orange | Optimum pH 7 Methyl orange was degraded at 69.23% after 240 min | Shinde et al. (2018) |
| Green material | Species | Composites | Properties | Potential applications | Ref |
|----------------|---------|------------|------------|-----------------------|-----|
| Plant          | Centaurea cyanus | Ag/Fe$_3$O$_4$/ZrO$_2$ | Particle sizes: 30–90 nm, saturation magnetization: 10 emu/g | Catalytic reduction of 4-nitrophenol and methyl orange, 3 cycles with swift reaction time (7.5–8 min) | Rostami-Vartooni et al. (2019) |
| Plant          | Justicia adhatoda | CeO$_2$/ZrO$_2$ | Nano-stick like structure (10–15 nm) Narrow band gap (3.37 eV) | Antimicrobial against *S. aureus* and *E. coli*, antioxidant ability, anti-biofilm | Pandiy et al. (2018) |
| Plant          | Hevea brasiliensis | Ni-doped ZrO$_2$ | Energy gap of 2.4–2.75 eV Narrow band gap (3.37 eV) | Nanoelectronics | Yadav et al. (2021) |
| Plant          | Leucas aspera | Sm-doped ZrO$_2$ (Sm$^{3+}$/ZrO$_2^{2+}$ = 3–11 mol.%) | Highly symmetric nanocubic (Fm-3 m) Band gap of 5.3–5.9 eV | High stable and reusability with six consecutive recycles | Gurushantha et al. (2016) |
| Plant          | Anacardium occidentale | Ag/ZrO$_2$ | Monoclinic and tetragonal phases of ZrO$_2$ Face-centered cubic crystal phase of Ag NPs (5–20 nm) on ZrO$_2$ surface | Not reported | Vivekanandhan et al. (2015) |
| Plant          | Commelina diffusa (-7-hydroxy-4´-methoxy-isoflavon from extract as a reducing and stabilizing agent) | Cu/ZrO$_2$ | Reduced agglomeration of CuZrO$_2$ with particle size of 18–25 nm Cu/ZrO$_2$ exhibited the good stability up to three days after the synthesis process | Catalytic reduction of 2,4-dinitrophenylhydrazine, various organic dyes such as congo red, nigrosin, methyl orange in the presence of NaBH$_4$ at room temperature Ultrafast reaction time for methyl orange (1 s), dinitrophenylhydrazine (40 s), and congo red (150 s) High recyclability at least 5 times | Hamad et al. (2019) |
| Plant          | Daphne alpine | V$_2$O$_5$/ZrO$_2$ | Surface area of 214 m$^2$/g, particle size of 41.74 nm, band gap 3.93 eV Thermal stability up to 1000 °C | The degradation efficiencies of V$_2$O$_5$/ZrO$_2$ against methyl orange (76.9%) and picloram (86%) for 75 min | Racheed et al. (2020) |
| Plant          | Ageratum conyzoides | Ag/ZrO$_2$ | Particle size of 50 nm Face centered cubic of Ag on ZrO$_2$ surface | The reduction of 2,4-dinitrophenylhydrazine, 4-nitrophenol, nigrosin and congo red High catalytic performance for reduction of 4-NP into 4-AP (100%) for 6 min, and 2,4-DAPH for 50 s | Maham et al. (2020) |
| Plant          | Aloe Vera | Mg-doped ZrO$_2$ | Shape: hollow microspheres, and tetragonal phase | ZrO$_2$–Mg (2 mol.%) gave the highest degradation efficiency (93%) Total organic carbon test gave the mineralization rate of 79% after 60 min of reaction Five consecutive cycle runs | Renuka et al. (2016) |
Antifungal

provide more insightful evidence into the antifungal activities of green zirconia nanoparticles. It is also recommended to better elucidate the underlying mechanisms of inhibiting the fungal cells by green ZrO₂ nanoparticles for future researches (Tables 4 and 5).

Anticancer

On average, one-third people are diagnosed with cancer in their lifetime, causing one of the world’s leading deaths (Sung et al. 2021). The evolution of cancer disease is the growth of abnormal cells to invade other tissue of the body. Many therapies such as surgery, chemotherapy, stem cell, and radiotherapy have been developed to treat this disease (Mooney et al. 2018). Bionanotechnology has recently played a vital role in designing novel anticancer agents and nanocarriers for drug delivery (Dai et al. 2017). Accordingly, nanoparticles can preferentially approach and accumulate in tumors thanks to the permeable effects, but this accumulation process is mostly influenced by physicochemical factors in the body (Beik et al. 2019). Green ZrO₂ nanoparticles have several emerging advantages such as nanoscale size, high surface area, stability, and good biocompatibility to overcome these barriers. On the tumor surface, green ZrO₂ nanoparticles bind to the macromolecules such as proteins and then penetrate into the tumor cells (Ranjii-Burachaloo et al. 2018). The chief mechanisms of cell damage by green ZrO₂ nanoparticles are not elucidated insightfully in the published literatures. According to Table 3, there are a limited number of works that evaluated the cytotoxicity activity of green ZrO₂ nanoparticles biosynthesized from Lagerstroemia speciosa leaf and Nephelium lappaceum fruit extracts against the breast cancer cell lines (Saraswathi and Santhakumar 2017; Isacfranklin et al. 2020). Therefore, many further investigations of anticancer activity of green ZrO₂ nanoparticles are required.
Environmental remediation

Removal of antibiotic drugs

Environmental pollution is currently presenting as one of the most globally critical issues, demanding many urgent actions (Crini and Lichtfouse 2019). In particular, the presence of emerging pollutants such as antibiotics can reduce the quality of water sources, and generate many potential threats for human and aquatic animals (Tahrani et al. 2015; Oberoi et al. 2019; Kovalakova et al. 2020). Several works reported the evolution of antibiotic resistance genes in rivers, hospital effluents, wastewater treatment plants, and sediments (Kairigo et al. 2020; Böger et al. 2021). Therefore, the treatment of antibiotic pharmaceuticals from water media by effective methods is necessarily required. Considering the non-degradable and chemically stable structure of antibiotics, adsorption may be the best remediation method due to its enormous advantages including high efficiency, fast kinetic, and tunable procedure (Tran et al. 2019b, 2020d, b; Dang et al. 2021; Nguyen et al. 2021e). However, the selection and development of novel advanced materials for water treatment are still challenging. Green ZrO\textsubscript{2} nanoparticles can meet the requirements for a good adsorbent since they obtain many outstanding properties. Indeed, biologically synthesized ZrO\textsubscript{2} nanoparticles have proved their superior functionality of the surface, stability, and eco-friendly synthesis, and hence, exhibiting their capacity for antibiotic adsorption (Table 4).

Silva et al. (2019) optimized the tetracycline removal efficiency of ZrO\textsubscript{2} nanoparticles biosynthesized from Euclea natalensis roots by response surface methodology. In this experiment, the conditions of ZrO\textsubscript{2} nanofabrication were set up \(2^3\) factorial design for extract content, precursor content, and pyrolysis temperature. The calcination temperature at 550 °C was found the most suitable. Under optimized removal conditions at initial concentration of 20 mg/L and adsorbent dose of 0.5 g/L, green ZrO\textsubscript{2} nanoparticles obtained the tetragonal phase with extremely small particle size (5.90–8.54 nm) and exhibited the best adsorptive capacity of 30.45 mg/g for tetracycline. However, this capacity value was relatively lower than that obtained by most adsorbents. To improve such situation, Debnath et al. (2020) investigated the synthesis of green ZrO\textsubscript{2} nanoparticles by bacterial community, specifically, Pseudomonas aeruginosa species for the treatment of tetracycline in aqueous solutions. Green ZrO\textsubscript{2} owned both monoclinic and tetragonal crystal phases with small size (6.41 nm). They optimized the operating tetracycline adsorption parameters such as pH 7 and contact time of 15 min to reach the very promising capacity of 526.32 mg/g. Importantly, green ZrO\textsubscript{2} could be easily regenerated with dilute NaOH solution and distilled water, bringing good reusability up to 5 cycles. The authors also proposed the major role of electrostatic interaction for their adsorption mechanisms. With the high performance of stability, reusability, and removal efficiency, green ZrO\textsubscript{2} nanoparticles can be ideal nanomaterials for water purification.

Removal of textile dyes

In addition to the contamination of antibiotics in water, many textile dyes are becoming persistent pollutants. It was estimated that a large amount of textile dyes is yearly discharged into water sources, causing many adverse impacts on human health (Zhou et al. 2019). They are also known to dismiss the solar light penetration, resulting in the reduction of photosynthesis efficiency as well as metabolism processes in several aquatic species (Tran et al. 2019a, 2020c). As same as antibiotics, most of these organic dyes are chemically stable, and difficult to degrade by biological methods such as phytoremediation (Nguyen et al. 2021f). However, another feasible solution is to apply the nanomaterials for the remediation of textile dyes through the adsorption and photocatalytic routes (Tran et al. 2020e; Nguyen et al. 2021c, a). Table 4 summarizes the main findings of recent publications on the treatment of dyes using green ZrO\textsubscript{2} nanoparticles.

By adsorption pathway, Alagarsamy et al. (2022) demonstrated the high efficiency of green ZrO\textsubscript{2} nanoparticles biosynthesized from the pericarp extract of Sapindus mukorossi for the removal of methylene blue. They optimized the adsorption conditions such as pH 10, ZrO\textsubscript{2} dose of 0.3 g/L, and 20 mg/L of tetracycline to remove 94% methylene blue for 300 min. Although the adsorption capacity was low (23.25 mg/g), this adsorbent could be reused for three consecutive cycles. In terms of catalytic degradation by green ZrO\textsubscript{2} nanoparticles, textile dyes were investigated more widely (Table 4). Take the study by Shinde et al. (2018) for example, both methylene blue and methylene orange dyes could be sufficiently degraded (61–91%) by green ZrO\textsubscript{2} nanoparticles biosynthesized from the Ficus benghalensis leaves under ultraviolet light irradiation. Moreover, the authors reported the catalytic optimal conditions at pH 7 and catalyst loading of 1.5 g/L. The plausible catalytic mechanism was suggested by the incorporation of reactive oxygen species such as ·O\textsubscript{2}−, and ·OH into the degradation process of dyes. However, carrying out the dyes degradation under ultraviolet light irradiation as mentioned by Shinde et al. (2018) may be unfavorable for practical applications. Al-Zaqri et al. (2021) improved this harsh input by using green ZrO\textsubscript{2} nanoparticles biosynthesized from Wrightia tinctoria leaves. The outcomes were very promising because this dye was mostly removed with the high first-order rate constant \(k_1 = 0.9837 \text{ min}^{-1}\) in the presence of sunlight irradiation. The plant extract may be contributed significantly to the formation of green ZrO\textsubscript{2} nanoparticles with smaller particle...
size (17 nm) as well as narrower bandgap energy (3.78 eV), which exhibited better catalytic performance.

**Removal of other pollutants**

The presence of other emerging pollutants such as fluoride can cause several severe diseases including skeletal fluorosis, teeth mottling, bones deformation, and many neurological issues (Affonso et al. 2020). The process of chemical fertilizing using hydrofluoric acid generates the amount of fluoride ions in water. It is therefore necessary to treat this pollutant before being discharged from the aqueous media. Prasad et al. (2014) assessed the adsorptive efficiency of adopting green ZrO2 nanoparticles from Aloe vera extract. Nearly 99% of F− ions were adsorbed ZrO2-based adsorbent, and fluoride chemisorption capacity was obtained at 96.58 mg/g. The authors also found the exothermic, spontaneous, and recyclicity nature of the fluoride adsorption process, suggesting the potential of green ZrO2 in fluoride decontamination. Although the high performance of ZrO2 nanoparticles produced from green extract sources for the treatment of antibiotics, textile dyes, and fluoride has been presented in the previous works, their great potentials for removing other emerging pollutants such as heavy metal ions, chlorinated compounds, persistent organic pollutants, pharmaceuticals, and personal care products have not yet exploited. It is recommended for further studies to explore the strength of green ZrO2 nanoparticles for the treatment of other emerging pollutants in the environmental remediation field.

**Other applications**

Silicon and many commercialized semiconductors-based solar cells have exhibited their highly efficient performance, but their production process is generally expensive (Roy et al. 2020). Moreover, these processes use chemically synthesized protocols, hindering sustainable development. To reach the purposes of green production, the modification of ZrO2 nanoparticles aims to suffer this major drawback. For example, Yadav et al. (2021) inserted an amount of transition metal nickel (Ni) into ZrO2 nanoparticles using rubber latex extract to obtain monoclinic, tetragonal, and cubic Ni-doped ZrO2 polycrystalline. The band gaps were very narrowed between 2.4 and 2.75 eV, making Ni-doped ZrO2 as potential nanoelectronic devices. Rasheed et al. (2020) reported the utilization of Daphne alpine leaf extract to synthesize green V2O5/ZrO2 nanocomposite. They not only observed the increase in surface area (214 m2/g) but also found the improvement of bandgap energy (3.93 eV) and thermal stability (1000 °C). It was suggested that V2O5 efficiently enhanced the electron separation, resulting in good photocatalytic activities against orange (76.9%) and picloram (86%) of green V2O5/ZrO2.

Doping other transition metals (Sm, Cu, Ag) significantly changes the electronic nature and crystalline phases of the
nanocomposites, leading to the enhancement of their stability, recyclability, and photocatalytic activities. Indeed, Gurushantha et al. (2016) fabricated the green ZrO2 with samarium dopant (Sm3+/ZrO2+ = 3–11 mol.%) using Leuca Aspera extract. Sm/ZrO2 composites with the doping of 11% dopant obtained highly symmetric nanocubics, and the highest photocatalytic degradation of acid green with six consecutive recycles in the presence of solar energy. This suggested that the Sm/ZrO2 composites are highly stable and reusable. Hamad et al. (2019) demonstrated the presence of 7-hydroxy-4´-methoxy-isoflavon from Commelina diffusa leaves reduced the agglomeration of Cu/ZrO2 nanoparticles (18–25 nm) with good stable ability during the post-synthesis. Moreover, the green Cu/ZrO2 nanoparticles catalyzed the reduction and degradation of 2,4-dinitrophenyl-hydrazine, and organic dyes including congo red, nigrosin, and methyl orange with ultrafast reaction times (1–150 s). In another study, Maham et al. (2020) obtained green Ag/ZrO2 nanocomposites (50 nm) using Ageratum conyzoides plant extract. The catalyst could reduce and degrade a range of substrates such as 2,4-dinitrophenylhydrzone, 4-nitrophenol, nigrosin, and congo red, indicating the greatly catalytic performance of green Ag/ZrO2 nanocomposite.

Although green ZrO2 nanoparticles exhibit their very high catalytic performance, the separation and recovery of these non-magnetically small-size nanomaterials from the aqueous post-reaction is disadvantageous. The intercalation of magnetic components (e.g., Fe3O4) into ZrO2 nanomaterials should be therefore required. Inspired by this idea, Rostami-Vartooni et al. (2019) have successfully the Centaurea cyanus-biofabricated Ag/Fe3O4/ZrO2 nanocomposites with average particle size between 30 and 90 nm. The nanocomposite could be easily separated from the reaction solutions because its saturation magnetization value was obtained at 10 emu/g. The authors also reported some good results of catalytic reduction of 4-nitrophenol and degradation of methyl orange green Ag/Fe3O4/ZrO2 with a swift reaction time between 7.5 and 8 min. Therefore, this advantage may urge the recyclability experiments of magnetic ZrO2 nanocomposites.

ZrO2-based nanocomposites biosynthesized from the green extract sources can offer a high diversity of morphology and open new applications. For example, Pandiyan et al. (2018) employed the green synthesis of CeO2 doped ZrO2 nano sticks with small particle size (10–15 nm) and bandgap (3.37 eV) from Justicia adhatoda leaf extract. This nanocomposite exhibited high antibacterial activities against S. aureus, and E. coli and high antioxidant activity. They also reported the application of CeO2/ZrO2 nano sticks as potential biofilms against many pathogeneses caused by bacteria such as S. marcescens. Renuka et al. (2016) successfully created the hollow structure of Mg-doped ZrO2 microspheres using Aloe Vera leaf extract. This nanocomposite exhibited high antibacterial activities against S. aureus, and E. coli and high antioxidant activity. They also reported the application of CeO2/ZrO2 nano sticks as potential biofilms against many pathogeneses caused by bacteria such as S. marcescens.
bio compounds such as polysaccharides from *Aloe Vera* leaf might chelate with $Zr^{4+}$ and $Mg^{2+}$ ions during the reaction of complexation. The burning calcination of these complexes allowed to acquire the hollow structure of Mg/ZrO$_2$ microspheres. They found the Mg/ZrO$_2$ composites with the magnesium doping rate of 2 mol% gave the highest photocatalytic activity against Rhodamine B (93%) in the presence of ultraviolet irradiation. Controlled oxygen vacancies on active sites may be attributable to the enhanced catalytic performance of green Mg/ZrO$_2$.

**Perspective**

Green synthesis of ZrO$_2$ nanoparticles from microbial and botanical sources has gained great attention because they offer many advantages such as eco-friendly and low-cost manufacturing compared with chemical synthesis methods. Green ZrO$_2$ nanoparticles possess many distinctive structure properties including small size, good dispersion, diverse surface chemistry, large surface area, and sufficient bandgap energy, making them extend excellent applications regarding biomedical and environmental fields. In many cases, the green ZrO$_2$ nanoparticles experimentally exhibit better antibacterial, antifungal, catalytic and adsorptive performance than chemically synthesized ZrO$_2$ nanomaterials. These superior prospects of green ZrO$_2$ nanoparticles may pay the way for developing advanced ZrO$_2$-based nanosensors for smart agriculture, high-efficiency solar cells for energy harvesting, and electronic nanodevices for many high-technological manufacturing industries.

Although green ZrO$_2$ nanoparticles hold many key advantages, there are still several limitations that need further investigation. *Firstly*, regarding the synthesis procedure for ZrO$_2$ nanoparticles microbial sources, the control of biomass growth rate, metabolite generation, and biochemical properties of cultivation environment should be investigated vigorously. For the botanical sources, the determination of bioactive compound ingredients in the plant extract should be conducted to better elucidate the formation mechanism of ZrO$_2$ nanoparticles. Moreover, the formation of green ZrO$_2$ nanoparticles should be optimized to obtain their expectable properties of particle size, charge nature, surface energy, and surface chemistry. *Secondly*, regarding the treatment of pollutants such as antibiotics, and textile dyes, the treatment of reused ZrO$_2$ nanoparticles has not been addressed. This action may generate secondary pollution; hence, it is necessary to solve the post-treatment problems. Moreover, most studies on the application of green ZrO$_2$ nanoparticles only reported their findings under laboratory modes, resulting in the difference between simulated and practical results. This drawback can be therefore addressed if the tests are conducted under practical conditions. *Thirdly*, to the best of our knowledge, the potential of green ZrO$_2$ nanoparticles is substantially unexploited in the many environmental fields, *e.g.*, the treatment of emerging persistent organic pollutants, pharmaceuticals and personal care products, non-steroidal anti-inflammatory drugs, and toxic gas detection. With the excellent properties of green ZrO$_2$ nanoparticles, it is expected that they will contribute significantly to the development of new environmental remediation approaches. **Ultimately**, the combination of dopants with green ZrO$_2$ nanoparticles can accelerate their inherent properties of the nanocomposites, expanding many promising applications. However, it is generally difficult to control the interactions of green extract with the dopants during the ZrO$_2$ based nanocomposites. The optimization of the biomass proportion should be evaluated to avoid these interactions. Even though the formation mechanism and application of green ZrO$_2$ based nanocomposites have been better understood in some published works, the number of researches is still limited. The knowledge of the overall interactions between ZrO$_2$ nanomaterials and the environment is just burgeoning. The profound evaluations of the ecotoxicity of green ZrO$_2$ nanoparticles should be continuously conducted to avoid possible future challenges.

**Conclusion**

The present review addressed some aspects of synthesis, properties, and potentials of green ZrO$_2$ nanoparticles and their nanocomposites for biomedical, environmental remediation, and other applications. The bioactive compounds derived from microbial and botanical extracts are attributable to the bioreduction, biocapping, biochelating, and bio-stabilization during the transformation of zirconium sources into green ZrO$_2$ nanoparticles. Through the structural characterization techniques, green ZrO$_2$ nanoparticles are found with many morphologies such as nanospheres, nanorods, nano chains. They possess diverse surface chemistry, high stability, and wide bandgap energy possibly due to the effect of green extracts. Some applications regarding the antibacterial, antifungal, anticancer, photocatalytic, and adsorptive activities of green ZrO$_2$ nanoparticles have been overviewed and discussed systematically. The advanced properties and applications of green ZrO$_2$-based nanocomposites were also mentioned. To orientate the future researchers, some discussions of knowledge gaps and prospects of green ZrO$_2$ nanoparticles have been clarified. The green ZrO$_2$ nanoparticles and their nanocomposites are highly expected to obtain many good results in various applications.
The authors declare that software application or standards.

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