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REVIEW

Green synthesis of ZrO_2 nanoparticles and nanocomposites for biomedical and environmental applications: a review

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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

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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 (ZrO_2) or zirconia is one of the highly stable oxides, created by thermalizing zirconium compounds (Hassan and Jalil 2022). Depending on the various synthesis routes, ZrO_2 can present in the crystalline phases involving monoclinic, tetragonal, and cubic (Zhang et al. 2018). Bulk ZrO_2 has a wide bandgap energy, typically ranging from 5.0 to 7.0 eV. Because of the ultrahigh stability and very low toxicity, ZrO_2 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 ZrO_2 an ideal nanomaterial, promoting the green strategy for synthesizing ZrO_2 nanoparticles.

In general, there are two major approaches to synthesize the ZrO_2 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 ZrO_2 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 ZrO_2 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 ZrO_2 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 ZrO_2 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 pollutions, biogenic synthesis of ZrO_2 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 ZrO_2 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 ZrO_2 nanoparticles and their nanocomposites. Therefore, the main objectives of this study are to discuss the current findings on green ZrO_2 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 ZrO_2 nanoparticles; (ii) structural characterizations of green ZrO_2 nanoparticles; (iii) biomedical, adsorption, catalytic, and other applications of green ZrO_2 nanoparticles; and (iv) emerging applications of green ZrO_2-based nanocomposites (Fig. 1). We briefly elucidate some challenging and prospective aspects of green ZrO_2 nanocomposites for expanding their potential applications.

Green synthesis of ZrO_2 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 ZrO_2 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 (Fariq et al. 2017). Underlying mechanism of forming ZrO_2 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,

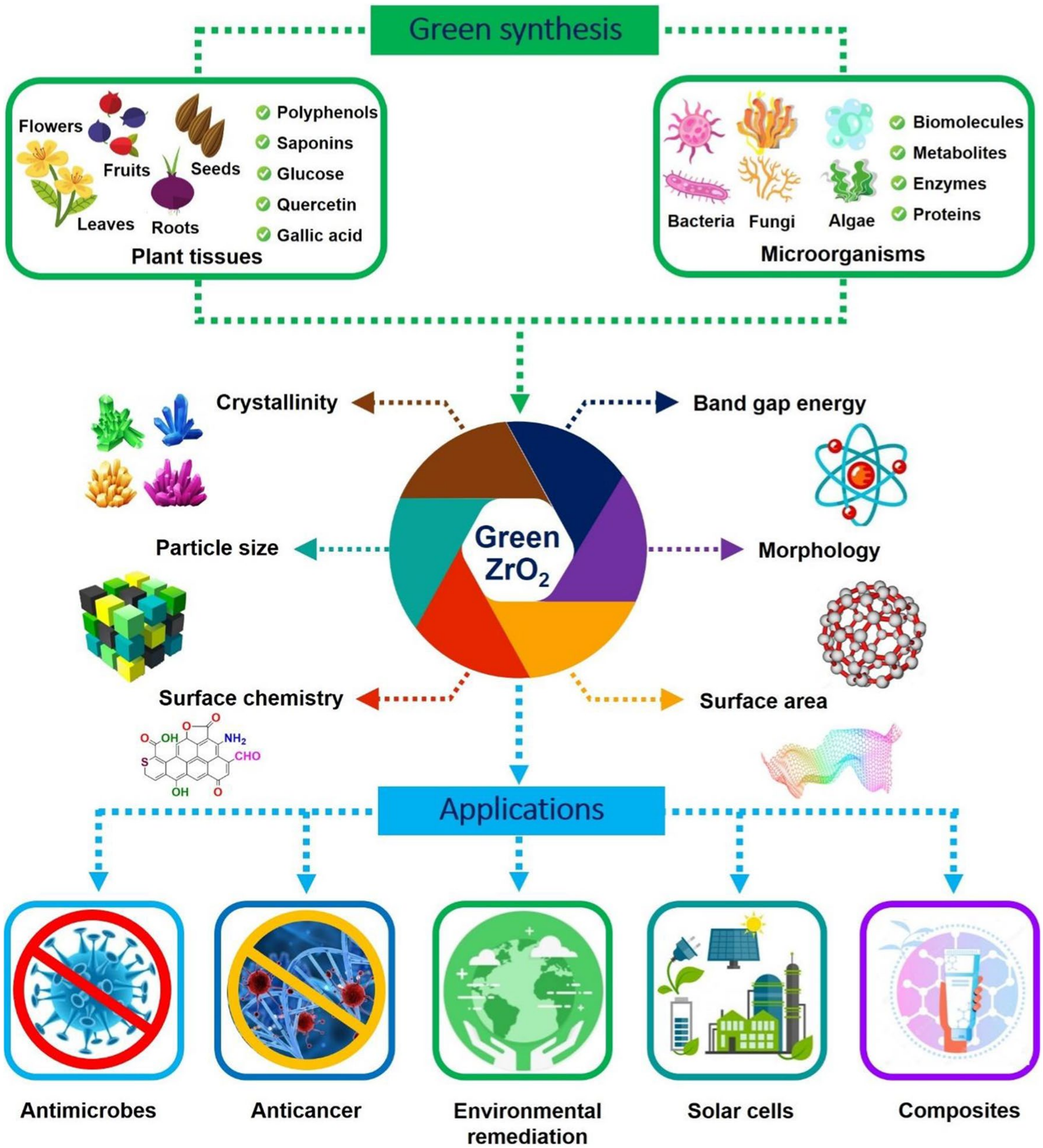

Fig. 1 Green synthesis of ZrO_2 nanoparticles and their applications in biomedical, adsorption, catalysis, and nanocomposite fabrication. The plant tissues including flowers, fruits, seeds, leaves, roots, etc. possess many phytochemicals such as polyphenols, saponins, quercetin, and gallic acid. Microorganisms including bacteria, fungi, algae, etc. can secrete biomolecules, metabolites, enzymes, and proteins. These phytochemicals and biomolecules participate in the green synthesis of ZrO_2

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 $ZrOCl_2{\cdot}8H_2O$ for incubation for 12 h. Interestingly, highly crystalline ZrO_2 nanomaterial was facilely precipitated under room-temperature condition (37 °C). Moreover, this 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 $ZrOCl_2 \cdot 8H_2O$ was addedto 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

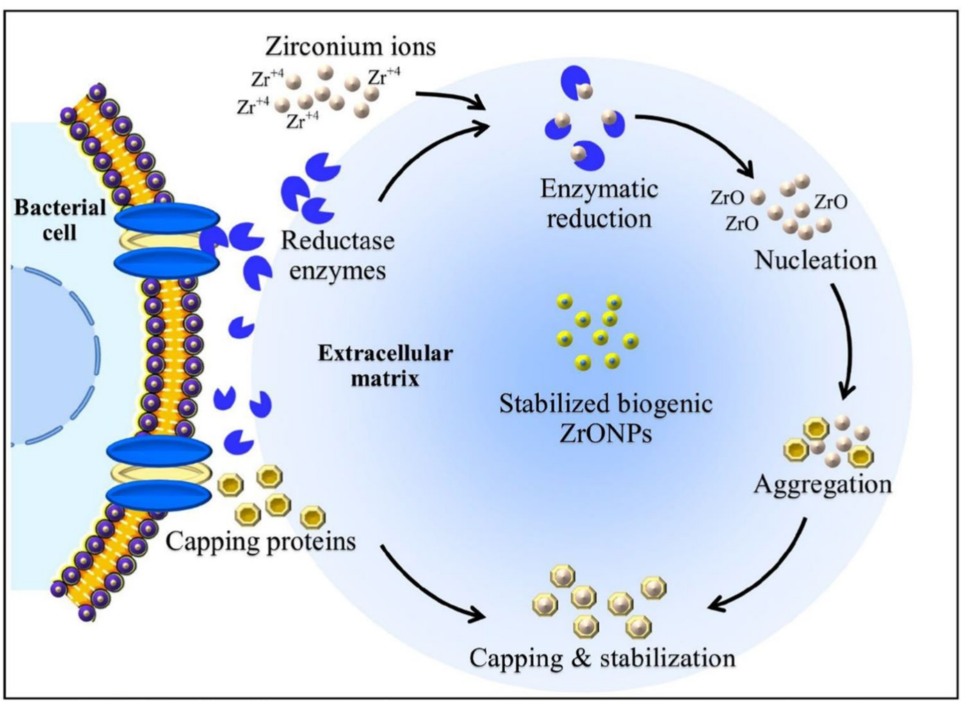

Fig. 2 Extracellularly biosynthesized mechanism of zirconia nanoparticles using *Enterobacter* sp. strain. Bacteria extracellularly secrete some specific enzymes such as reductase, which take responsibility for enzymatic bioreduction of Zr^{4+} ions. The underlying mechanism is attributable to the redox of nicotinamide adenine dinucleotide (NAD^+/NADH), which supplies electrons to the reduction of Zr^{4+} ions during nucleation. Capping proteins can also be released into the 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 ZrO_2 than other biological synthesis methods such as bacteria and plants.

Ghomi et al. (2019) reported that the formation of ZrO_2 by extracellular secretion of *Penicillium* fungal species including *P. aculeatum, P. notatum, and P. purpurogenome.* Under the optimum conditions at pH 9, and 1.5 mM Zr^{4+}, the solution color changed from pale yellow into deep yellow after seven-day incubation, affirming the generation of colloidal ZrO_2. In terms of the ZrO_2 biofabrication mechanism, the enzymes presenting in the fungal supernatants may aid to transform Zr^{4+} ions into ZrO_2 nanoparticles. Bansal et al. (2004) cultured another fungal species named *Fusarium oxysporum* for the successful biosynthesis of ZrO_2 nanoparticles from the ZrF_6^{2-} source at room temperature. To gain insight into the major role of proteins secreted from this fungus for the extracellular hydrolysis of ZrF_6^{2-}, the resulting filtrates were lyophilized to test for hydrolytic activity. The findings confirmed the cationic nature of proteins to bind easily to ZrF_6^{2-} anions, which could not be found in other fungal species (*e.g., C. lunata, C. gloeosporioides, Phomopsis sp., A. niger*). More importantly, ZrF_6^{2-} anions showed a non-toxicity to *Fusarium oxysporum*, opening the enormous prospects for large-scale biosynthesis of ZrO_2 nanoparticles. To sum up, the highlighted eco-friendly, biocompatible, and energy-conserving advantages of the fungal biosynthesis of ZrO_2 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 ZrO_2. Indeed, Kumaresan et al. (2018) successfully bioprepared tetragonally nanostructured ZrO_2 from the aqueous extract of seaweed *Sargassum wightii*. The biosynthesis procedure was tunable with the grind of $ZrO(NO_3)_2{\cdot}H_2O$ with *Sargassum wightii* extract within 20 min. ZrO_2 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 ZrO_2 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 ZrO_2. It is also necessary to elucidate the underlying mechanisms for the formation of ZrO_2 nanoparticles using algal species.

Plants

Plants are the most prevalent, locally available, highly cost-effective resource for biosynthesis of ZrO_2 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 ZrO_2 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 ZrO_2 nanoparticles is considerably accelerated in comparison with microbial routes. The bioreduction of Zr^{4+} 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 ZrO_2 production process. *Thirdly*, the synthesis of ZrO_2 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 ZrO_2 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 ZrO_2 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 ZrO_2 nanoparticles (Fig. 3b). The phytochemicals participate in the formation and stabilization of octahedral complex of Zr^{2+}–phytochemicals. The presence of phytochemicals also aids to hamper the aggregation of ZrO_2 nanoparticles during thermal calcination. This may boost the surface area of ZrO_2 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,

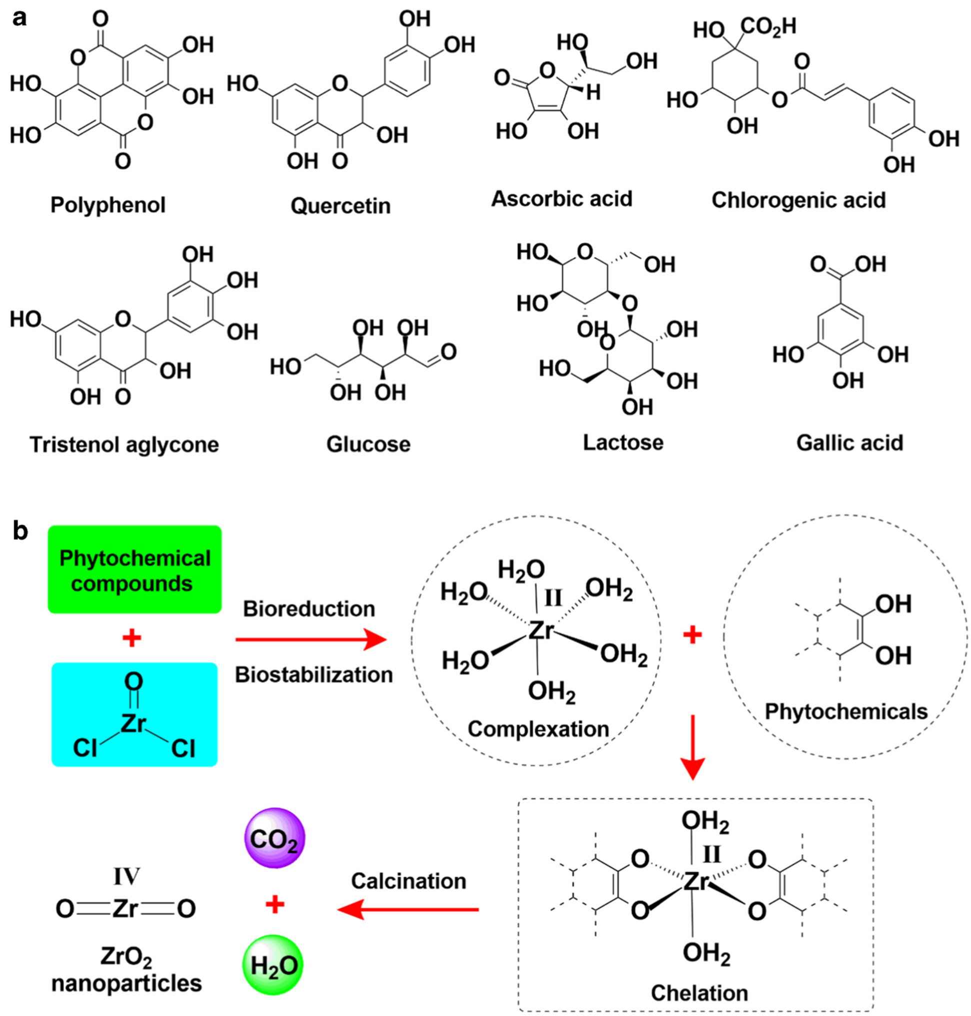

Fig. 3 Phytochemicals in plant extracts (**a**), and proposed mechanism for the biosynthesis of ZrO_2 nanoparticles using these phytochemicals (**b**). Phytochemicals act as bioreductants to convert Zr^{4+} into octahedral complex $[Zr(H_2O)_6]^{2+}$. This enhances the stability and hinders the clustering of zirconium complexation during chelation with phytochemicals Zr^{2+}–phytochemicals. The calcination of Zr^{2+}–phytochemicals complex produces ZrO_2 nanoparticles in tandem with the release of CO_2, H_2O, 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.
Plant species	Plant tissue	Zr source	Zr/extract ratio (v/v)	Temperature	Time	Temperature	Time	
Euclea natalensis	Roots	$ZrOCl_2$	1:1	Not reported	3 h	550 °C	3 h	Silva et al. (2019)
Aloe vera	Leaves	$ZrOCl_2$	Not reported	Room	4 h	500 °C	Not reported	Gowri et al. (2014)
Salvia rosmarinus	Leaves	$ZrOCl_2$	1:4	70 °C	1 h	600 °C	2 h	Davar et al. (2018)
Sapindus mukorossi	Pericarp	$ZrOCl_2$	1:1	60 °C	3–4 h	500–700 °C	Not reported	Alagarsamy et al. (2022)
Wrightia tinctoria	Leaves	$ZrOCl_2$	1:1	75 °C	3–4 h	800 °C	Not reported	Al-Zaqri et al. (2021)
Ficus benghalensis	Leaves	$ZrOCl_2$	1:1	Microwave	15 min	500 °C	3 h	Shinde et al. (2018)
Moringa oleifera	Leaves	$ZrOCl_2$	1:1	60 °C	3–4 h	700–800 °C	Not reported	Annu et al. (2020)
Helianthus annuus	Seeds	$ZrOCl_2$	1:5	Not reported	2–3 h	600 °C	4 h	Goyal et al. (2021)
Tinospora cordifolia	Leaves	$ZrOCl_2$	10:1	55 °C	40 min	Not reported	Not reported	Joshi et al. (2021)
Nephelium lappaceum	Fruit	$Zr(NO_3)_4$	7:1	180 °C	18 h	500 °C	4 h	Isacfranklin et al. (2020)
Nyctanthes arbor-tristis	Flower	$ZrOCl_2$	2:1	Not reported	2 h	300–500 °C	3 h	Gowri et al. (2015)

expensive chemical antioxidants or reductants, and shortening the number of complicated synthesis steps.

The general botanical biosynthesis of ZrO_2 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 ZrO_2 products. The former is carried out under the incorporation of plant extract into zirconium source, mainly $ZrOCl_2 \cdot 8H_2O$. The proper temperature and time for forming Zr^{2+}–phytochemicals are 55–75 °C, and 1–4 h, respectively. This step can also be assisted by microwave irradiation (900 W) to accelerate the complexation rate up to 15 min (Shinde et al. 2018), or by alkaline addition (Joshi et al. 2021). The Zr^{2+}–phytochemicals complex is totally separated by centrifugation or paper filtration (Gowri et al. 2014). The latter experiences the thermal calcination at high temperature (500–800 °C) for 2–4 h to convert this complex into ZrO_2 nanoparticles.

Properties of green ZrO_2 nanoparticles

Crystallinity

Bulk ZrO_2 nanoparticles exist in three common crystalline phases involving monoclinic, tetragonal, and cubic. Biologically synthesized ZrO_2 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 ZrO_2 nanoparticles bioproduced from *Salvia Rosmarinus* plant extract with the reflecting planes at (111), (200), (220), (311), (222), and (400). Another study indicated the monoclinic phase of ZrO_2 synthesized from *Helianthus annuus* plant extract (Goyal et al. 2021). However, tetragonal phase is the most prevalent single-crystal phase of green ZrO_2 nanoparticles. Green ZrO_2 nanoparticles can also have multiphase crystallinity, which includes at least two phases (Sathishkumar et al. 2013; Debnath et al. 2020; Isacfranklin et al. 2020). The presence of crystal phase in green ZrO_2 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 ZrO_2 phase.

Optical bandgap

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 (Makuła et al. 2018). It can be determined from diffuse reflectance spectra. While the bulk ZrO_2 nanoparticles have a wide bandgap energy (~5.0 eV), green ZrO_2 nanoparticles exhibit values ranging from 3.7 to 5.5 eV (Table 2). Indeed, Goyal et al. (2021) fabricated ZrO_2 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 ZrO_2 nanoparticles biosynthesized from *Wrightia tinctoria* leaf extract. They also gave an assumption about more contribution from external surface defects ZrO_2 nanoparticles. Meanwhile, obtaining higher bandgap energies (5.4–5.5 eV) of other green ZrO_2 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 ZrO_2 nanoparticles acquire sufficient gap energies, capable of exhibiting their excellent catalytic and biomedical activities through the formation of reactive oxygen species.

Particle size

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 ZrO_2 nanoparticles. From the database of previous studies (Table 2), the particle size of ZrO_2 nanoparticles is distributed from 5 to 150 nm, commonly less than 50 nm. Compared with chemical approaches, biological methods generally create ZrO_2 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 ZrO_2 nanoparticles (Kumaresan et al. 2018; Silva et al. 2019; Debnath et al. 2020). Small-size ZrO_2 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 ZrO_2 nanocrystals biosynthesized from biological routes can be explored by scanning/transmission electron microscopy analysis. Accordingly, green ZrO_2 nanoparticles can present a wide range of morphological properties such as nanospheres (mainly), nanochains,

Table 2 Properties of green ZrO_2 nanoparticles

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, carbonyl, carboxylate	Kumaresan et al. (2018)
Bacterium	*Enterobacter sp.*	33–75	Nanosphere	Not reported	Not reported	Hydroxyl, carbonyl, 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	Debnath et al. (2020)
Fungus	*Penicillium*	< 100	Nanosphere	Not reported	Not reported	Hydroxyl, carbonyl, amine	Ghomi et al. (2019)
Fungus	*Fusarium solani*	Not reported	Nanosphere	Tetragonal	Not reported	Hydroxyl, carbonyl	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, carbonyl, zirconium hydroxide	Gowri et al. (2014)
Plant	*Curcuma 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, carbonyl, 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, carbonyl, amine	Saraswathi and Santhakumar (2017)
Plant	*Ficus benghalensis*	14.7	Nanosphere	Monoclinic, tetragonal	4.9	Hydroxyl, alkyl, carbonyl	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, alkene, amide	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, carbonyl	Joshi et al. (2021)
Plant	*Nephelium lappaceum*	50	Nanorod	Monoclinic, cubic	Not reported	Hydroxyl	Isacfranklin et al. (2020)
Plant	*Nyctanthes arbor-tristis*	< 150	Nanoflake	Tetragonal	5.31	Not reported	Gowri et al. (2015)

nanorods, semi-nanospheres, nano-sized ovals, and nanoflakes (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,

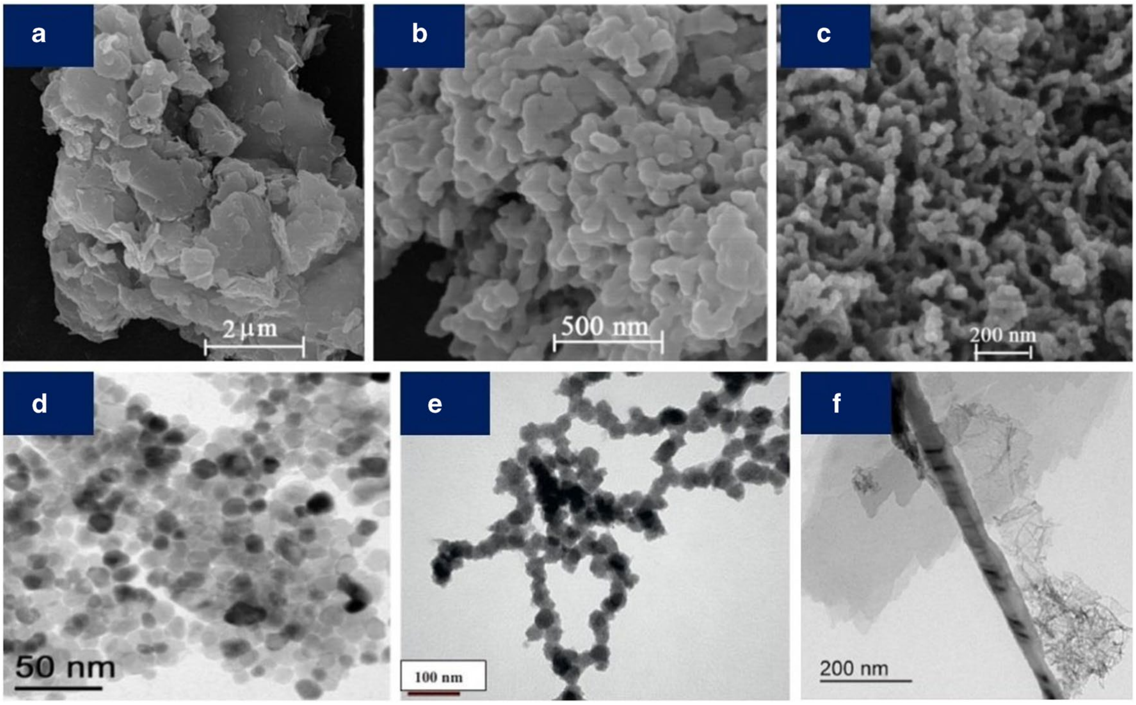

Fig. 4 Field emission scanning electron microscope photographies of ZrO_2 nanoparticles produced at 600 °C without *Rosmarinus officinalis* leaf extract (**a**), and from *Rosmarinus officinalis* leaf extract with the ratio between the extract and zirconium source 3:1 (**b**), and 6:1 (**c**). The role of plant extract is to improve the dispersion of ZrO_2 nanoparticles during biosynthesis. Reproduced and adapted with the permission of Elsevier from reference (Davar et al. 2018). Transmission electron microscopy photographs of ZrO_2 nanospheres (**d**), nano chains (**e**), and nanorods (**f**) biosynthesized from *Acinetobacter* sp. bacterial community (Suriyaraj et al. 2019), *Curcuma longa* tuber extract (Sathishkumar et al. 2013), and *Nephelium lappaceum* fruit extract (Isacfranklin et al. 2020), respectively. Reproduced and adapted with the permission of Elsevier from references (Sathishkumar et al. 2013; Suriyaraj et al. 2019; Isacfranklin et al. 2020)

Helianthus annuus seeds, *Tinospora cordifolia* leaves, and *Laurus nobilis* leaves (Table 2). Meanwhile, other ZrO_2 morphologies are relatively rare, mostly synthesized by plant sources. For example, Sathishkumar et al. (2013) obtained ZrO_2 nano-chains by the hydrolysis of ZrF_6^{2-} in the presence of *Curcuma longa* tuber extract. Gowri et al. (2015) also successfully biosynthesized ZrO_2 nanoflakes by the hydrolysis of $ZrOCl_2{\cdot}8H_2O$ using aqueously soluble carbohydrates extracted from *Nyctanthes arbor-tristis* flower. Semi-spherical and oval ZrO_2 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 ZrO_2 morphologies can bring many benefits for practical applications. As an example, (Isacfranklin et al. 2020) indicated that ZrO_2 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 ZrO_2 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 ZrO_2 nanoparticles may be originated from the phytochemical compounds. Considering the calcination of the complex of Zr^{2+}–phytochemicals, almost all organic components are decomposed into volatiles or simple products such as CO_2, N_2, and H_2O. 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 H_2O-adsorbed surface of ZrO_2 (Kumaresan et al. 2018; Silva et al. 2019; Annu et al. 2020). The functionalization of ZrO_2 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 ZrO_2 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 ZrO_2 nanoparticles during the thermolysis. This enhances the surface area and the number of active sites of ZrO_2 nanoparticles. For example, Shinde et al. (2018) reported the surface area (88 m^2/g) of ZrO_2 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 ZrO_2 to methylene blue (91%) and methyl orange (69%) for 240 min. These results of green ZrO_2 were remarkably higher than those obtained by the chemical methods such as anodization in H_2O_2/NH_4F/ethylene glycol electrolyte (Rozana et al. 2017), decomposition of $Zr(OH)_4$-urea complex (Sudrajat et al. 2016), polymer-assisted sol–gel synthesis (Dhandapani et al. 2016).

Stability

Compared with chemically produced nanomaterials, biologically produced ZrO_2 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 ZrO_2 nanoparticles can be assessed by the zeta potential measurements. Basically, the zeta potential values are different with zero, normally minimum $\pm$ 30 mV, indicating the high stability of ZrO_2 nanoparticles (Jameel et al. 2020). Indeed, Suriyaraj et al. (2019) demonstrated the well-dispersed, anti-clustering, and nanostructured ZrO_2 suspension by the measurement of zeta potential (36.5 $\pm$ 5.5 mV). Chau et al. (2021) also measured the high zeta potential ($-$32.8 mV) of ZrO_2 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 ZrO_2 nanoparticles

Biomedical applications

Biomedical compatibility of green ZrO_2 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 effectivity may restrain the biomedical application of these nanomaterials. Green ZrO_2 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 ZrO_2 nanoparticles.

Antibacterial activity

The plasma membrane of bacteria is mostly constituted of proteins (*e.g.,* peptidoglycan macromolecules) that charge negatively (Li et al. 2019). ZrO_2 nanoparticles charge positively on their surface, leading to electrostatic interactions with bacterial membranes (Xing et al. 2018). This facilitates the biosorption and bioaccumulation of ZrO_2 nanoparticles on the cell walls. With the nanoscale size, high surface area, and good biocompatibility of green ZrO_2 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.,* $\cdot O_2^-$, and $\cdot OH$) is the underlying mechanism (Akintelu and Folorunso 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 ZrO_2 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 ZrO_2 biosynthesized from *Nyctanthes arbor-tristis* plant extract. The

Table 3 Biomedical applications of green ZrO_2 nanoparticles

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), and *S. aureus* (15 mm) *for* ZrO_2 NPs, and those against *E. coli* (30 mm), and *S. aureus* (22 mm) for ZrO_2-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 ZrO_2 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-Zaqri 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. pneumonie*)	Zone of inhibition against *E. coli* (13 mm), *P. aeruginosa* (13.5 mm), *S. aureus* (12.0 mm), and *K. pneumonie* (12.5 mm) at a ZrO_2 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 ZrO_2 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)

Table 3 (continued)

Green material	Species	Type of activity	Target	Main findings	Ref
Plant	*Nephelium lappaceum*	Anticancer	Human breast cancer cell lines (MCF-7)	Half-maximum inhibitory concentration of 55.32 μg mL^{-1}	Isacfranklin et al. (2020)
Plant	*Lagerstroemia speciosa*	Anticancer	Breast cancer cell lines (MCF-7)	The cells viability with 18% inhibition was observed at 500 μg/mL of zirconia nanoparticles Nearly 30–40% of the cells showed blebbing	Saraswathi and Santhakumar (2017)

inhibition zones were used to measure the antibacterial activity of green ZrO_2 nanoparticles. The values were significantly improved in the case of the cotton fabric treated with ZrO_2 nanoparticles. With very low green ZrO_2 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 ZrO_2 nanoparticles, *e.g.,* small size particles and high surfaces, enhancing their antibacterial activities. In addition to plant extract, ZrO_2 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 ZrO_2, 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 ZrO_2 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 ZrO_2 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.,* $\cdot O_2^-$, and ·OH) and free radicals (Akintelu and Folorunso 2020). With the high biocompatibility, green ZrO_2 nanoparticles can pervade through the fungal cell membranes by endocytosis. In the intracellular environment, ZrO_2 nanoparticles account for detrimental functions such as ribosomal, chromosomal, and mitochondrial damages through the formation of reactive oxygen species (Fig. 5). Moreover, ZrO_2 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 ZrO_2 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 ZrO_2 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 ZrO_2 (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 ZrO_2 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

Table 4 Environmental remediation applications of green ZrO_2 nanoparticles

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 ZrO_2-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 (k_1)	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 Santhakumar (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)

Table 5 Potential applications of green ZrO_2-based nanocomposites

Green material	Species	Composites	Properties	Potential applications	Ref
Plant	*Centaurea cyanus*	$Ag/Fe_3O_4/ZrO_2$	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)	Antibacterial against *S. aureus*, and *E. coli*, antioxidant ability, anti-biofilm	Pandiyan et al. (2018)
Plant	*Hevea brasiliensis*	Ni-doped ZrO_2	Energy gap of 2.4–2.75 eV	Nanoelectronics	Yadav et al. (2021)
Plant	*Leucas aspera*	Sm-doped ZrO_2 (Sm^{3+}/ZrO^{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 High sunlight-driven degradation of Sm/ZrO_2 with 11 mol.% dopant against acid green dye	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 Cu/ZrO_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- dinitrophenilhydrazine, 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), dinitrophenilhydrazine (40 s), and congo red (150 s) High recyclability at least 5 times	Hamad et al. (2019)
Plant	*Daphne alpine*	V_2O_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_2O_5/ZrO_2 against methyl orange (76.9%) and picloram (86%) for 75 min	Rasheed 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 At least 5 cycles for reusability study	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)

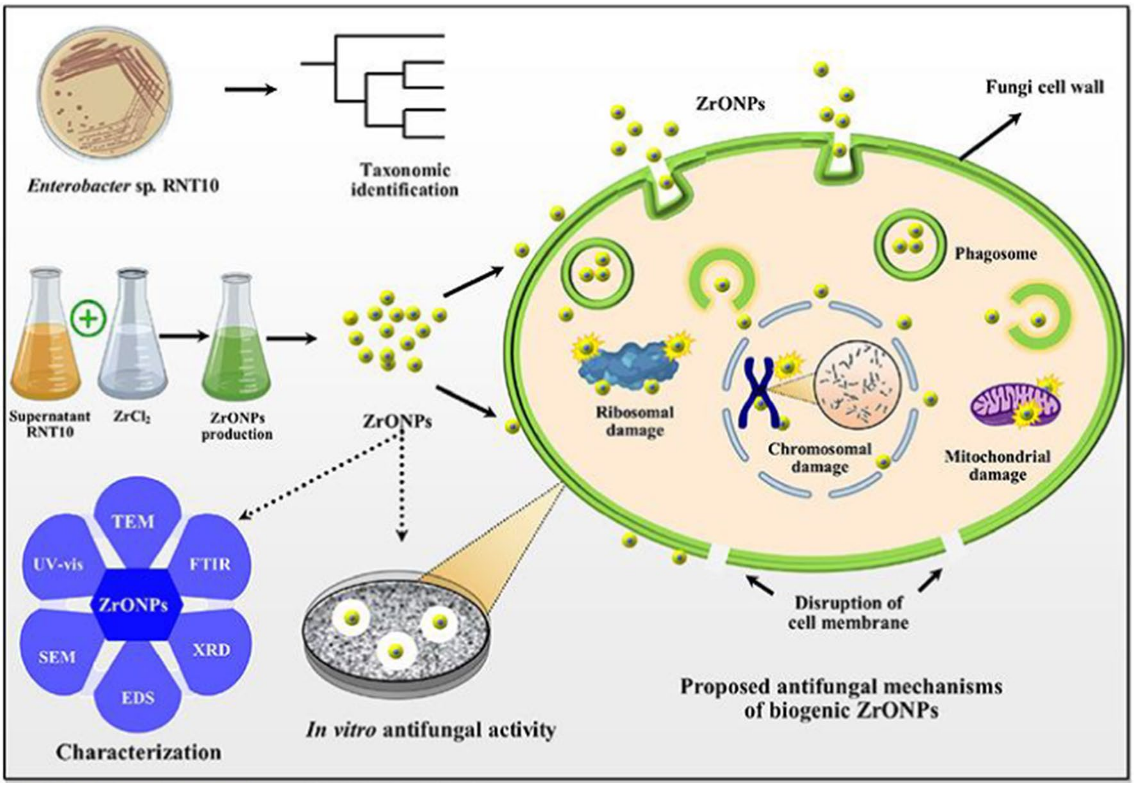

Fig. 5 Synthesis, characterization, and proposed antifungal mechanisms of green zirconia nanoparticles. Green zirconia nanoparticles may pervade through the fungal cell membranes, causing many detrimental effects on ribosomal, chromosomal, and mitochondrial parts. They may change the intracellularly physicochemical properties to cause cell stress and break the integrity of fungal cells. Reprinted with the permission of Elsevier from reference (Ahmed et al. 2021). Abbreviations: TEM, transmission electron microscope; FTIR, Fourier-transform infrared spectroscopy; XRD, X-ray diffraction; EDS, energy-dispersive X-ray spectroscopy; SEM, scanning electron microscopy; UV–Vis, ultraviolet–visible spectroscopy; ZrONPs, zirconia nanoparticles; RNT10, an isolated strain of *Enterobacter*

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 ZrO_2 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 ZrO_2 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 ZrO_2 nanoparticles bind to the macromolecules such as proteins and then penetrate into the tumor cells (Ranji-Burachaloo et al. 2018). The chief mechanisms of cell damage by green ZrO_2 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 ZrO_2 nanoparticles biosynthesized from *Lagerstroemia speciose* 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 ZrO_2 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_2 nanoparticles can meet the requirements for a good adsorbent since they obtain many outstanding properties. Indeed, biologically synthesized ZrO_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_2 nanoparticles biosynthesized from *Euclea natalensis* roots by response surface methodology. In this experiment, the conditions of ZrO_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_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_2 nanoparticles by bacterial community, specifically, *Pseudomonas aeruginosa* species for the treatment of tetracycline in aqueous solutions. Green ZrO_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_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_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_2 nanoparticles.

By adsorption pathway, Alagarsamy et al. (2022) demonstrated the high efficiency of green ZrO_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_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_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_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 $\cdot O_2^-$, 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_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 ($k_1 = 0.9837$ min^{-1}) in the presence of sunlight irradiation. The plant extract may be contributed significantly to the formation of green ZrO_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 serve 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 ZrO_2 nanoparticles from *Aloe vera* extract. Nearly 99% of F^- ions were adsorbed ZrO_2-based adsorbent, and fluoride chemisorption capacity was obtained at 96.58 mg/g. The authors also found the exothermic, spontaneous, and recyclability nature of the fluoride adsorption process, suggesting the potential of green ZrO_2 in fluoride decontamination. Although the high performance of ZrO_2 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 ZrO_2 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 biological synthesis of low-cost dye-sensitized solar cells should be of great interest (Alami et al. 2019). Green ZrO_2 nanoparticles can become one of the prospective electronic nanomaterials for solar energy harvesting. Several studies reported the biosynthesis of green ZrO_2 nanoparticles for solar cell applications. For example, Majedi et al. (2016) synthesized the cubic-phase zirconia nanoparticle with small particle size (21 nm) using lemon juice. They reported the electrical conductivity and activation energy of ZrO_2 nanoparticles of 0.0034 S/cm, and 31.015 kJ/mol, respectively. These results exhibited a relatively electronic performance of green ZrO_2 as potential electrolytes. Vennila et al. (2018) also have investigated the synthesis of ZrO_2 using *Gloriosa Superba* tuber powder and found the potential to create high-efficiency electrodes. However, the applications of green ZrO_2 nanoparticles as solar cells are still limited. The reaction mechanism, as well as the ways to improve the energy harvesting efficiency, are sufficiently unexplored. It is recommended that further studies should be investigated.

ZrO_2-based nanocomposites

The incorporation of dopants can considerably change the properties of ZrO_2 nanoparticles in terms of particle size, uniformity, electronic nature, and morphologies. Green synthesis of ZrO_2-based nanocomposites allows improving some disadvantages of bandgap energy, surface area, surface chemistry, and stability of inherent ZrO_2 nanoparticles. To reach a green and sustainable approach, the synthesis of ZrO_2-based nanocomposites uses microbial or botanical sources, which play a key role in the bioreduction, biocapping, biochelating, and biostabilizing process for the transformation into composites. For example, Vivekanandhan et al. (2015) functionalized the surface of ZrO_2 nanocrystallites with silver nanoparticles using *Anacardium occidentale* leaf extract (Fig. 6). The bio compounds from this extract might participate in the bioreduction of Ag^+ into Ag and nucleation of Ag nanospheres (5–20 nm) on ZrO_2 surface. The authors also reported the inhibition of the deformation of Ag/ZrO_2 nanocomposites in the larger presence of leaf extract. This phenomenon may be attributable to swift bioreduction of Ag^+ ions before their dispersion on the surface of ZrO_2. Therefore, a sufficient amount of plant extract is important to obtain the structure of ZrO_2-based nanocomposites. Table 5 summaries several potential applications of green ZrO_2-based nanocomposites.

Because ZrO_2 nanoparticles exhibit a wide bandgap (~ 5.0 eV), the absorption of visible light is difficult, hampering their photocatalytic and nanoelectronic applications. The modification of ZrO_2 nanoparticles aims to suffer this major drawback. For example, Yadav et al. (2021) inserted an amount of transition metal nickel (Ni) into ZrO_2 nanoparticles using rubber latex extract to obtain monoclinic, tetragonal, and cubic Ni-doped ZrO_2 polycrystalline. The band gaps were very narrowed between 2.4 and 2.75 eV, making Ni-doped ZrO_2 as potential nanoelectronic devices. Rasheed et al. (2020) reported the utilization of *Daphne alpine* leaf extract to synthesize green V_2O_5/ZrO_2 nanocomposite. They not only observed the increase in surface area (214 m^2/g) but also found the improvement of bandgap energy (3.93 eV) and thermal stability (1000 °C). It was suggested that V_2O_5 efficiently enhanced the electron separation, resulting in good photocatalytic activities against orange (76.9%) and picloram (86%) of green V_2O_5/ZrO_2.

Doping other transition metals (Sm, Cu, Ag) significantly changes the electronic nature and crystalline phases of the

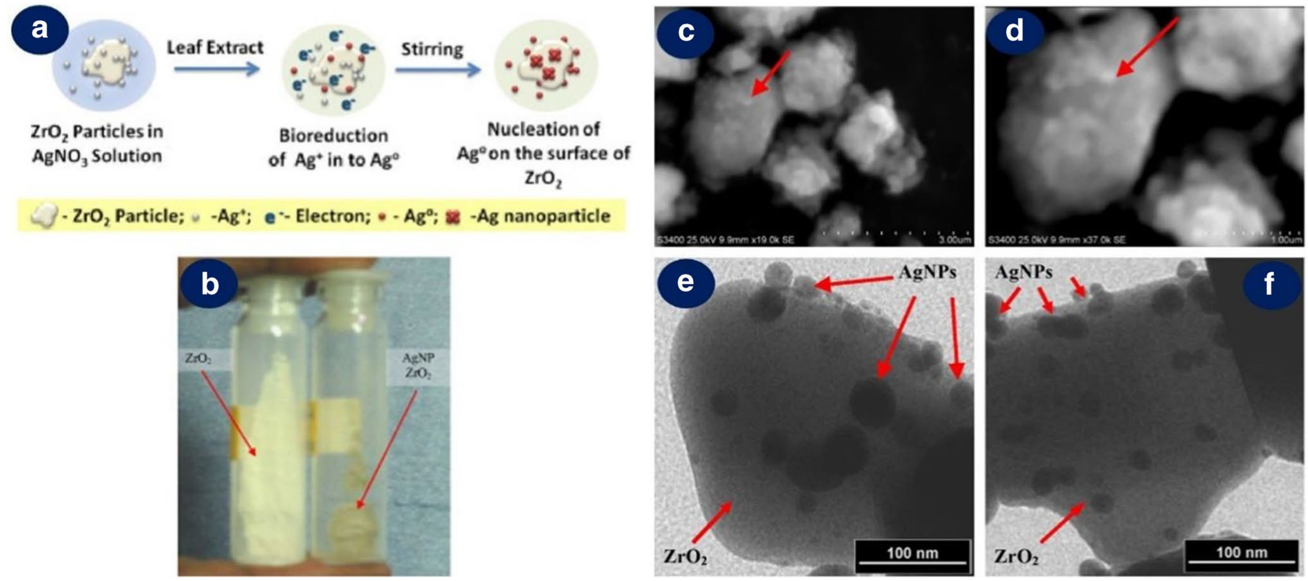

Fig. 6 The bioreduction of Ag^+ into Ag and nucleation of on ZrO_2 surface for the biosynthesis of green Ag/ZrO_2 nanocomposites using *Anacardium occidentale* leaf extract (**a**); the photograph of as-synthesized green ZrO_2 and Ag/ZrO_2 nanocomposites (**b**); scanning electron microscopy (**c**, **d**) and transmission electron microscopy (**e**, **f**) microphotographies of green Ag/ZrO_2 nanocomposites. The presence of leaf extract efficiently leads to the good dispersion and high biostabilization of Ag nanospheres (5–20 nm) on the surface of ZrO_2. However, larger addition of leaf extract inhibits the formation of Ag/ZrO_2 nanocomposites. Reproduced and adapted with the permission of Elsevier from reference (Vivekanandhan et al. 2015). Abbreviations: AgNP, silver nanoparticles

nanocomposites, leading to the enhancement of their stability, recyclability, and photocatalytic activities. Indeed, Gurushantha et al. (2016) fabricated the green ZrO_2 with samarium dopant (Sm^{3+}/ZrO^{2+} = 3–11 mol.%) using *Leucas Aspera* extract. Sm/ZrO_2 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/ZrO_2 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/ZrO_2 nanoparticles (18–25 nm) with good stable ability during the post-synthesis. Moreover, the green Cu/ZrO_2 nanoparticles catalyzed the reduction and degradation of 2,4-dinitrophenylhydrazine, and organic dyes including congo red, nigrosin, methyl orange with ultrafast reaction times (1–150 s). In another study, Maham et al. (2020) obtained green Ag/ZrO_2 nanocomposites (50 nm) using *Ageratum conyzoides* plant extract. The catalyst could reduce and degrade a range of substrates such as 2,4-dinitrophenylhydrazine, 4-nitrophenol, nigrosin, and congo red, indicating the greatly catalytic performance of green Ag/ZrO_2 nanocomposite.

Although green ZrO_2 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.,* Fe_3O_4) into ZrO_2 nanomaterials should be therefore required. Inspired by this idea, Rostami-Vartooni et al. (2019) have successfully the *Centaurea cyanus*-biofabricated $Ag/Fe_3O_4/ZrO_2$ 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 $Ag/Fe_3O_4/ZrO_2$ with a swift reaction time between 7.5 and 8 min. Therefore, this advantage may urge the recyclability experiments of magnetic ZrO_2 nanocomposites.

ZrO_2-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 CeO_2 doped ZrO_2 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 CeO_2/ZrO_2 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 ZrO_2 microspheres using *Aloe Vera* leaf extract. They assumed that bio compounds such as polysaccharides from *Aloe Vera* leaf might chelate with Zr^{4+} and Mg^{2+} ions during the formation of Mg/ZrO_2 microspheres. The authors assumed that 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, nonsteroidal 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 biostabilization 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.

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