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PHOTOCATALYTIC PROPERTIES AND APPLICATION OF TiO_2 AND ZnO NANOPARTICLES

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Abstract: In this review article, the photocatalytic properties and applications of TiO_2 and ZnO nanoparticles were studied. Special attention has been paid to their role in advanced oxidation processes. Key aspects of their electronic structure, energy gap, and crystal modifications were discussed, explaining how these factors can affect photocatalytic performance. The advantages and limitations of TiO_2 and ZnO were studied in detail. Special attention was devoted to their practical application in the degradation of pharmaceuticals, pesticides, and dyes, underlying their environmental significance. The review also provides a brief overview of the development of next-generation photocatalysts based on TiO_2 and ZnO, highlighting the need for improved efficiency and stability.

Keywords: photocatalysts; advanced oxidation processes; environmental application; nanomaterials; degradation of pollutants

1. Introduction

Advanced oxidation processes (AOPs) utilize oxidants to break down organic pollutants such as pharmaceuticals, pesticides, dyes, microplastics, etc. by synthesizing highly reactive radicals, for instance, hydroxyl radicals ($^{\bullet}OH$) [1]. These pollutants are oxidized and ultimately transformed into smaller molecules, such as carbon dioxide (CO_2) and water (H_2O). In photocatalytic processes, titanium dioxide (TiO_2) and zinc oxide (ZnO) are most often employed as photocatalysts under solar or UV irradiation, which results in the cleavage of mighty $^{\bullet}OH$ [2]. These radicals are potent [3] in the breakdown of organic material, and the process is eco-friendly because TiO_2/ZnO are nontoxic and stably persistent [4]. These processes proved to be economical as well as cost-effective [5].

Heterogeneous photocatalysis is an AOPs that has received considerable attention in recent years, especially for application in water disinfection and purification. TiO_2 is the most used photocatalyst in water treatment processes because of its low cost and toxicity, relatively high efficiency, and high stability (chemical and thermal) [6]. More recent

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developments indicated that the efficiency of photocatalytic processes with TiO_2 is substantially improved in the presence of some inorganic oxidants because of additional radical formation [7]. UV light generates the most efficient photocatalytic degradation, yet it can also lead to the generation of undesired byproducts since it can be harmful and expensive to use [8]. Though less efficient, solar light offers a more environmentally friendly substitute with less chance of undesired side effects. Although successful in specific applications, solar light generally exhibits reduced effectiveness in photodegradation relative to UV light due to its inability to regulate the light spectrum and the diminished concentration of UV radiation, which is essential for numerous photocatalytic activities [9].

Nanomaterials represent particles of external size approximately 1–100 nm [10]. These materials are very attractive for many applications because of their improved chemical reactivity and physical properties such as optical absorption, thermal conductivity, and mechanical strength. The nanoscale dimension provides an improved way to make nanomaterials attractive for various applications. TiO_2 and ZnO in nanoscale form exhibit multiple advantages over traditional forms [11]. The enhanced photocatalytic activity of ZnO and TiO_2 nanostructures has recently attracted considerable interest. Forms of nanostructures such as nanoparticles, nanorods, and nanotubes have higher specific surface areas, which increases the reactive sites for photocatalytic reactions [12]. The photocatalytic efficiency is also enhanced due to the ease of separable surface charge carrier interactions, which decreases the recombination rate. The range of applications is broadened in photocatalysis, environmental detoxification, and solar energy conversion due to nanostructures, which can easily undergo doping and modification of surface functional groups. In addition, co-precipitation, hydrothermal, and solvothermal methods based on sols, gels, and other biological systems enable the modification of TiO_2 and ZnO nanostructures with targeted properties, while minimizing the use of hazardous chemicals, thus increasing the sustainability of the processes. With all these factors, it is apparent that compared to the bulk materials, the nanostructures are more efficient, nontoxic, and cost-effective [11].

Compared with the individual constituents, i.e., ZnO nanorods and amorphous TiO_2 nanoparticles, the TiO_2-ZnO hybrid nanostructures exhibited increased catalytic performance [13]. The drastic suppression of the green emission observed in the photoluminescence of TiO_2-ZnO hybrid nanostructures indicates an excellent process of charge transfer and separation. Within the nanohybrid system [14], ZnO offers an active surface along with emissions due to charge transfer, and TiO_2 induces electron-hole separation, both leading to photocatalytic efficiency. Further, the particular interfacial areas and geometric orientation between TiO_2 and ZnO also play a role in enhancing photocatalysis [15].

Therefore, the primary objective of this review was to provide a detailed comparative analysis of the photocatalytic properties of TiO_2 and ZnO nanoparticles. Special focus was on

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their structural, electronic, and functional characteristics that affect their photocatalytic performance. By examining their mechanisms of action, energy bandgap structures, and crystal modifications, the review aims to point out the advantages and limitations of each material. In addition, the degradation of organic pollutants such as pharmaceuticals, pesticides, and dyes using these photocatalysts was discussed. Finally, the review seeks to identify key research gaps and offer perspectives on the future development of more efficient and stable TiO_2 and ZnO-based photocatalysts.

2. Photocatalytic process

The idea of photocatalysis has been around since 1839, but it was not until the late 1960s that more research was done on it, thanks to people like Boddy, Honda, and Fujishima [16]. People are becoming more interested in photocatalysis these days because it could help with sustainable development, and it could enable the storage of solar energy in chemicals, which are called solar fuels and artificial photosynthesis. In research, photocatalysis is the process of speeding up a photoreaction by using a catalyst. Pollutants that build up in the air, soil, and water, mainly because of human activity, are not removed from the environment because natural degradation processes are slow. Photocatalysis is used to speed up the process of removing them from the air, water, and other places [17].

The photocatalysis process starts when the photocatalyst absorbs light and produces electron-hole pairs. Since no negative basic charge is cleared out behind when an energized electron is extricated from a covalent bond, the location obtains a positive charge. The resultant hole, or emphatically charged location, remains within the valence band, whereas the energized electron moves to the conduction band [18]. These charge carriers move to the catalyst surface after being isolated, where they take part in redox responses, in which electrons decrease the accessible substances and holes oxidize them [19].

The method loses proficiency in case electrons (e^-) and holes (h^+) recombine as well rapidly. To make strides in photocatalytic execution, materials that ensure compelling charge partition and transport must be synthesized [20]. Beneath light with vitality possibly over its bandgap of 3.2 eV, TiO_2, a photocatalyst, becomes activated. An electron from the valence band moves to the conduction band, breaking through the bandgap [21]. Because of this, a hole remains within the valence band and acts as an oxidizing agent by taking electrons from the substances that must be oxidized, like water, which can create $OH^•$. In contrast [22], the excited state of TiO_2 molecules is unstable and tends to be stabilized by transferring the excited electrons to a reducing material such as oxygen (O_2) and creating superoxide radicals ($^•O_2^-$). With the use of oxidative organic pollutants, these reactive radicals ($OH^•$ and $^•O_2^-$) can promote efficient photocatalytic degradation of toxic substances [23]. The mechanism of the photocatalytic process using TiO_2/ZnO is shown in Figure 1.

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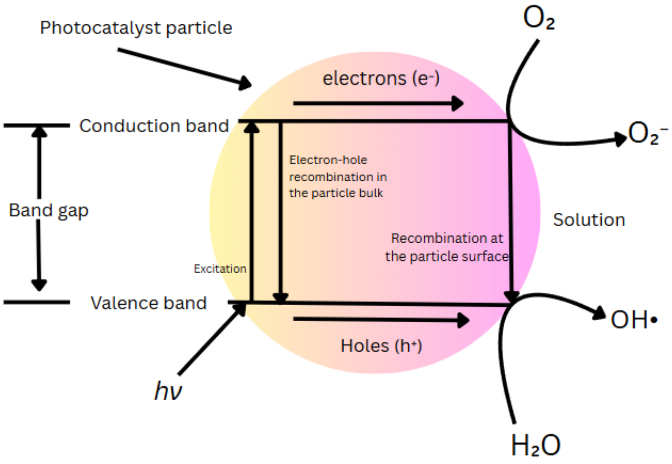

Figure 1. Photocatalytic process: Position of valence and conduction bands during TiO_2/ZnO excitation

2.1. Photocatalytic properties of TiO_2

TiO_2 is a nontoxic, highly stable, cost-efficient, and biodegradable semiconductor. It exhibits unique physicochemical properties such as outstanding optical properties, high refractive index, dielectric constant, chemical inertness, wide band gap, photostability, and resistance to corrosion. It can be activated under UV irradiation [24]. Fujishima discovered TiO_2 photocatalytic properties in 1972 [23]. Nowadays, TiO_2 has attracted substantial research interest, owing to its single-electron electronic structure, which gives it its photocatalytic activity. TiO_2 has a wide band gap, which can be tuned with different modifications of this material [25].

The electronic band structure of TiO_2 plays a crucial role in its photocatalytic activity. Among the wide variety of TiO_2 nanostructures reported [26], the most stable ones are three polymorphs: anatase, rutile, and brookite. Rutile has a direct band gap of 3.18 eV and an indirect band gap of 3.03 eV. For anatase, the direct band gap is 3.40 eV while the indirect band gap is 3.19 eV. Herein, the electronic band gap is indirect. However, for brookite, the direct band gap is 3.41 eV while the indirect band gap is 3.43 eV, suggesting that the electronic band gap is direct [27]. Anatase (tetragonal) has lattice parameters $a = 3.79$ Å and $c = 9.51$ Å, rutile (tetragonal) has $a = 4.59$ Å and $c = 2.96$ Å, while brookite (orthorhombic) has $a = 9.18$ Å, $b = 5.45$ Å, and $c = 5.14$ Å. Anatase is the most photocatalytically active polymorph, while rutile is the most thermodynamically stable. Comparison of the band structures reveals that anatase is more suitable for bandgap engineering and improving photocatalytic efficiency.

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2.2. Photocatalytic properties of ZnO

ZnO has gained substantial interest over the past decades owing to wide-ranging applications in catalysis, coatings, optics, electronics, photonics, photochemistry, optoelectronics, and magnetics. It represents a promising candidate for practical photocatalytic applications due to its excellent optical and chemical stability, low cost, and high photodegradation efficiency [28]. ZnO crystallizes predominantly in two crystal structures: hexagonal wurtzite and cubic zinc blende. Wurtzite phase constitutes the thermodynamically stable ground state for bulk ZnO crystals, while the metastable cubic zinc blende phase can be stabilized through epitaxial growth techniques [29]. The wurtzite phase is stable at atmospheric pressure, while the cubic zinc blende structure is stable at high pressures [30].

ZnO exhibits photocatalytic properties, with a bandgap between 3.3 eV and 3.4 eV and a wurtzite crystal structure. Its considerable binding energy (~60 meV) enables room temperature operation, providing robustness for various applications [31]. With a band gap of 3.3 eV, ZnO has gained huge attention due to its distinctive efficiency and low cost. Many efforts have been made to enhance the visible-light-driven photocatalytic properties of ZnO through doping, dye sensitization, ion implantation, coupling with narrow band gap semiconductors, etc. [32]. The intrinsic properties of ZnO are significantly influenced by point defects such as interstitial zinc, interstitial oxygen, oxygen vacancy, and zinc vacancy. These defects can change the electronic structure and participate in the recombination processes of electron-hole pairs. [33].

3. Comparison between TiO_2 and ZnO

In the mesoporous TiO_2 and ZnO nanostructures, the photocatalytic reaction rates and degradation pathways differ significantly. When photocatalytic reaction parameters remain equal, and the catalyst is changed, the photocatalytic activity of ZnO exhibits a higher rate of byproducts formation, whereas TiO_2 favors the direct degradation of organic compounds [34]. Oxygen readily adsorbs on TiO_2 surfaces, which facilitates the creation of surface defects and promotes direct degradation of organic compounds [35]. The goal of photocatalytic degradation includes detoxification and mineralization of organic pollutants. Therefore, the reaction rates are an important indicator of photocatalytic activity [36]. Photocatalytic reaction rates in the degradation of organic pollutants in wastewater have been compared for TiO_2 and ZnO [37]. ZnO appears to be somewhat more photocatalytically active compared to TiO_2 [34].

The photodegradation efficiency of ZrO_2/TiO_2 was higher compared to TiO_2 and increased with the number of atomic layer deposition cycles up to 45, beyond which it

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slightly decreased [38]. TiO_2/ZnO nanoparticles synthesized by rapid combustion featured similar structures and sizes of polycrystalline hexagonal wurtzite phase due to Ti^{4+} substitution at Zn^{2+} sites. N_2 adsorption/desorption analysis confirmed mesoporous, non-rigid aggregation structures for both ZnO and Ti-ZnO. The TiO-doped sample exhibited more than twice the surface area of undoped ZnO. Photocatalytic degradation kinetics of methylene blue had a rate constant of $2.54 \cdot 10^{-3}$ 1/min for Ti-ZnO compared to pure ZnO, which had a rate constant of $1.40 \cdot 10^{-3}$ 1/min. In the degradation of methylene blue, the dominant degradation pathway for TiO_2 involves the degradation of the C-N bond, whereas ZnO cleaves both the central C-N bond and aromatic rings [39].

In Table 1 we added tabular comparison of the TiO_2 and ZnO properties.

Table 1. Comparison of the properties of TiO_2 and ZnO nanostructures

Property	**TiO_2**	**ZnO**
Band gap (eV)	3.0 (rutile), 3.2 (anatase)	3.3
Refractive index	2.9	2.0
Dielectric Constant	~80–170	~8.5–10.2
Photocatalytic activity	High; lower rate of byproduct formation; favors direct degradation of organic compounds.	High; higher rate of byproduct formation; degrades both the central C–N bond and aromatic rings.
Oxygen adsorption	Oxygen readily adsorbs, facilitating surface defects and direct degradation.	Less pronounced oxygen adsorption.
Crystal Structures	Polymorphs anatase, rutile, brookite.	Polycrystalline hexagonal wurtzit, zincblende.
Specific surface area	30-200 m^2/g	10-100 m^2/g
Toxicity	Low	Low
Dominant degradation pathway	Degradation of the C–N bond.	Cleavage of the central C–N bond and aromatic rings.
Stability	High chemical and thermal stability.	Relatively stable, but with higher byproduct formation.
Synthesis methods	Sol–gel, hydrothermal, solvothermal, chemical vapor deposition, flame spray pyrolysis, and atomic layer deposition.	Rapid combustion, solvothermal, sol–gel, hydrothermal, precipitation, vapor-phase, microwave-assisted.
Applications	Water treatment, disinfection, photocatalysis, sunscreens.	Water treatment, disinfection, photocatalysis, solar cells.

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4. Strategies for improving the photocatalytic activity of TiO_2 and ZnO

Several strategies have been developed to enhance the photocatalytic activity and the visible light absorbance by TiO_2 and ZnO materials. Most often used strategies for improving photocatalytic activity of TiO_2 and ZnO include doping, coupling with other semiconductors, surface modifications, as well as other enhancement techniques [40].

Doping introduces impurity atoms into a semiconductor to create internal trapping states or shift the band edges, thereby improving photocatalytic performance. Metals frequently used as dopants are Ag, Cr, Cu, Fe, Co, V, Mn, Mo, Ni, Pt, Ti, and W. Non-metal dopants include B, C, F, I, N, and S [41]. The main aim of introducing a dopant is to create impurity energy levels within the bandgap of the semiconductor. Depending on the energy position, these states can increase visible light absorption and serve as either charge carrier traps or recombination centers [42].

Metal ion doping has emerged as a prominent method to improve the photocatalytic activity of TiO_2 and ZnO, especially under visible-light irradiation [43]. Incorporating various metal ions, such as Fe and Zn, enhances the degradation of organic pollutants beyond the performance of pure TiO_2 and ZnO because photogenerated electrons are effectively trapped by dopant ions, thereby reducing the rate of electron-hole recombination. For example, Fe doping narrows the bandgap and promotes charge separation, leading to an increase in photocatalytic activity in phenol degradation under visible light [44]. Other metals, including Rh, Pd, Pt, Ag, Au, Ni, Cr, W, Cu, and non-metal ions like N, have also been widely studied as dopants for TiO_2 and ZnO. Metal dopants with a smaller ionic radius prefer substitutional sites replacing cations, while larger ions tend to occupy interstitial sites within the lattice. In both cases, the band structure and surface activity are positively affected [37]. Metal ion doping can induce individual energy levels within the bandgap or alter the optoelectronic properties depending on the dopant species, concentration, and oxidation state. Doping systems with bi- or tri-metallic doping show synergistic effects that significantly improve the charge separation efficiency. On the other hand, the efficiency of co-doping with Fe and Zn is limited. It was shown that doping with one metal gives superior activity to doping with two of them. The influence of metal dopants on photocatalytic efficiency could be very specific, and it depends on the dopant type, characteristics of the host material, and the deposition techniques used [45].

Significant strategy for improving the photocatalytic efficiency of TiO_2 and ZnO is their doping with non-metallic elements, such as N, S, and F. Doping of TiO_2 and ZnO leads to the narrowing of the bandgap and the absorption of visible light [46]. This is the consequence of the new electronic states or modifying existing electronic states near the valence or conduction bands. Incorporation of N into mesoporous TiO_2 (mp-TiO_2) substantially extends its photocatalytic activity in the visible region. N doping shifts the optical absorption edge

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due to the formation of localized states and also inhibits crystal growth, helps retain the mesostructure, increases surface area, and provides sites for efficient charge separation. In mp-TiO_2, co-doping with non-metal elements can lead to many visible light-enhancing mechanisms, therefore improving the materials photocatalytic performance [47]. Doping with S includes the substitution of O with S in the TiO_2 lattice. This substitute results in a narrowed band gap and improved visible light absorption. Similarly, fluorine doping increases surface acidity and electron density, which promotes charge separation and visible-light responsiveness [48]. Non-metal–doped TiO_2 samples commonly exhibit enhanced degradation efficiency of organic dyes such as methylene blue, methyl orange, and rhodamine B under visible-light irradiation, a behaviour also observed in doped ZnO. Apart from improved visible light absorption by non-metallic dopants, they can also influence improved photocatalytic activity by slowing crystal growth, preserving mesostructures, and facilitating charge separation. Despite these advances, further improvements in light absorption, charge separation, and transfer processes are necessary to achieve the full potential of non-metal–doped TiO_2 and ZnO for practical photocatalytic applications [49].

Heterojunction *coupling* and *Z-scheme systems* represent two strategies for combining TiO_2 and ZnO with other semiconductors. Both approaches use different mechanisms to increase visible-light response and reduce charge recombination [50]. Coupling TiO_2 or ZnO with narrow bandgap semiconductors forms a type II heterojunction. At the interface, the energy levels of the conduction and valence bands drive transfer of photogenerated carriers so that electrons are accumulated on one side while holes are accumulated on the other, enhancing separation and extending spectral response. However, a high Schottky barrier and significant potential difference reduce carrier reduction capacity [51]. On the other hand, in the Z-pattern, two semiconductors are coupled with aligned conduction and valence bands in order to maximize redox capability. Further, the photogenerated electrons and holes are transported through a solid mediator. Z-pattern heterostructures are more complex to prepare, but in this way, the photocatalytic performance of materials is significantly improved [52].

An effective way to improve the photocatalytic activity of TiO_2 and ZnO is the formation of heterojunctions. Typically, a heterojunction is composed of two components. In the Type-II configuration, photogenerated electrons migrate into the conduction band component with the lower energy, whereas holes move to the higher-energy valence band material [53]. In contrast, Z-scheme heterojunctions unify complementary semiconductors of appropriately aligned bands in configurations that promote the removal of the low-potential electrons and the high-potential holes. This mechanism conserves the substantial reduction and oxidation potentials of the original uncombined semiconductors and, thus, sustains superior photocatalytic efficacy. Several TiO_2-based Z-scheme heterojunctions have been reported; these include NiO/TiO_2 [54], Bi_2O_3/TiO_2 [55], and WO_3/TiO_2 [56]. Common

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challenges for the realization of multilayer heterointerfaces encompassing electrolysis applications lie in the interface stability and the associated resistance.

Construction of a Z-scheme photocatalytic system that efficiently separates charge carriers, extends absorption to the visible region, and maintains strong redox capabilities represents a highly promising strategy to enhance photocatalytic activity. In such systems, two different semiconductors are integrated in a manner that suppresses charge recombination and ideally combines their redox properties [57]. When two semiconductors are coupled to form a standard type-II heterojunction, the photogenerated electrons in the conduction band of the semiconductor with the higher conduction band edge transfer to that with the lower conduction band edge, while photogenerated holes in the valence band of the semiconductor with the lower valence band edge transfer to that with the higher valence band edge. This results in both electrons and holes residing in bands with lower redox potentials, weakening the overall oxidation and reduction capacities [58].

Surface modifications include nanostructuring and coating of TiO_2 and ZnO. Nanostructures contribute spheres, sheets, flowers, rods, wires, tubes, cubes, and octahedra. Nanostructuring enhances photocatalytic activity by increasing the surface area-to-volume ratio, generating more active sites for reactions, and improving light absorption properties [59]. Nanostructuring additionally reduces recombination of photogenerated charge carriers by shortening their migration distance to active sites, thus increasing reaction efficiency. Coatings may be carbon-based, metals, or organics. Surface coating stabilizes high-energy crystal facets, optimizing charge transfer by directing photogenerated carriers to desired active sites at the particle surface while suppressing electron-hole recombination. Surface coating also extends light absorption to longer wavelengths by limiting electron-hole recombination in both TiO_2 and ZnO. orZnO nanostructures and surface coatings exhibit combined benefits from structural and electronic effects, including larger surface area, more sites for reaction, and improvements in light absorption with more efficient charge separation [60].

5. Application of TiO_2 and ZnO

The extensive use of pharmaceuticals, including analgesics, antibiotics, antipyretics, anti-inflammatory drugs, lipid regulators, and antiepileptics, has led to their continuous discharge into aquatic environments via wastewater systems. Many of these compounds are biologically active and can induce physiological changes at low concentrations, prompting investigations into their removal from wastewater and drinking water [61]. Photocatalytic degradation using TiO_2 and ZnO has been widely applied to pharmaceuticals. Systems containing the TiO_2/ZnO were exposed to UV or sunlight to generate hydroxyl radicals and other reactive oxygen species. The reactive oxygen species then react with the

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pharmaceutical compounds adsorbed on the catalyst surface, breaking the organic molecules down into various intermediates before complete mineralization to CO_2, H_2O, and inorganic ions [40].

Pesticides include organochlorines, organophosphates, pyrethroids, and carbamates. They are natural or synthetic substances used to control pests in agriculture and for public health problems. They pose environmental risks and require effective methods for the removal of their remains. Processes such as UV/H_2O_2 were efficient in the photocatalytic degradation of phenolic compounds and pesticides [62].

The synthetic dyes used in many industries, such as paper, textile, leather, and plastic products dyeing, have wide characteristics, such as azo, indigoid, oxazine, anthraquinone, acridine, and triarylmethane [63]. The dyes are not only toxic but also usually carcinogenic, highly visible, and biodegradable. These dyes can not be removed entirely from wastewater using the conventional process. Because of these problems, the efficacy of both TiO_2 and ZnO for removing dyes using the advanced oxidation process was examined, wherein they showed high efficiency and mineralization of selected dyes [64].

To provide a more structured overview of TiO_2 and ZnO photocatalytic performance, we have included a comparative table 2 summarizing relevant studies.

Table 2. Comparison of TiO_2 and ZnO for the degradation of different pollutants

Catalyst	Pollutant	Radiation type	Reaction time (min)	Degradation efficiency (%)	Reference
TiO_2	Metoprolol	UV	30 min	100%	[65]
ZnO	Metoprolol	UV	60 min	98%	[66]
TiO_2	Trimethoprim	UV	100 min	90%	[67]
ZnO	Trimethoprim	UV	20 min	95%	[67]
TiO_2	Ciprofloxacin	UV	90 min	88%	[68]
ZnO	Ciprofloxacin	UV	90 min	85%	[69]
TiO_2	Methylene Blue	UV	150 min	59%	[70]
ZnO	Methylene Blue	UV	150 min	100%	[70]
TiO_2	Congo Red	Sunlight	30 min	98%	[71]
ZnO	Congo Red	UV	80 min	100%	[72]
TiO_2	Rhodamine B	UV	40 min	100%	[73]
ZnO	Rhodamine B	UV	70 min	98%	[74]

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6. Conclusions

TiO_2 and ZnO nanomaterials constitute the basis of many of the next-generation applications in photocatalysis in the environmental and energy sectors. This review focuses on their properties and applications. Properties such as vast bandgap energies and surface morphology illustrate these materials' effectiveness as photocatalysts, affording them the capability to address environmental issues. The next generation of TiO_2 and ZnO would be developed to be used as self-cleaning surfaces, antimicrobial coatings, and molecular sensing. To realise the potential of TiO_2 and ZnO as the basis of a cleaner and energy-sustainable environment, the challenges and limitations faced by these materials must be identified, with an emphasis on future directions to steer their use in commercial and larger-scale operations.

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