

Photocatalytic Degradation of Antibiotics

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Various methods, including carbon filtration, ozonation, catalytic membranes, Fenton-like catalysis, sorption, and biodegradation have been employed to eliminate such enduring antibiotic substances. As a result, wastewater accumulates effluents from corporations, clinics, and farmland. Sewage treatment is typically regarded as the preferred method for treating these antibiotics. Photocatalysis has gained a lot of interest as an effective method for eliminating antibiotic contaminants since it is inexpensive, effective, and environmentally benign as it eliminates antibiotics in sunlight and under ambient conditions. The reactions endured by a semiconductor and the potential to absorb photons with energies higher than its energy band gap is called "photocatalytic degradation".

Keywords: antibiotics ; photodegradation ; ZnO-based nanoparticles ; mechanisms

1. Antibiotic Degradation Utilizing Pristine ZnO Photocatalyst

ZnO has multiple benefits because of its antigen-free, antimicrobial, antitumor, and wound-healing characteristics [1]. ZnO is an affordable material with minimal technical aspects, rendering it the more beneficial choice compared to TiO₂ as a photocatalyst. ZnO has a significant exciton binding energy of 60 meV and good capacity to absorb UV irradiation. ZnO exhibited better photocatalytic performance than TiO₂ due to its photogenerated electron-hole pairs, mobility, and isolation [2].

Effective photodegradation of ciprofloxacin (CIP) was exhibited by ZnO nanoparticles synthesized using an ethanolic root extract of Japanese knotweed with specific operational conditions, as shown in **Table 1**. A UV-vis spectrophotometer at 271 nm was used to analyze the level of CIP while the degradation process was being carried out under ultraviolet light ($\lambda_{\max} = 365$ nm). It is necessary to highlight that the λ_{\max} of CIP changed from 272 to 265 nm from the start to completion of degradation, emphasizing the disturbance of the chromophoric conjugation network. The perfect nanoparticle size of 14 nm had a perceived rate constant (k_{app}) of 0.038 min⁻¹ and completely degraded CIP during exposure for 100 min [3]. The degradation of CIP in water solutions with varied pH values under exposure to UV light was examined with ZnO nanoparticles synthesized using a chemical precipitation method. After 60 min of exposure, the maximal recorded CIP antibiotic degradation efficacy was around 18% at pH 4, 42% at pH 7, and 50% at pH 10. Pseudo-first order kinetics govern the photocatalytic degradation of ciprofloxacin. The free hydroxyl ions interact with holes (h⁺) and generate hydroxyl radicals (OH[•]) that possess strong oxidizing potential, thereby enhancing the photocatalytic degradation of ciprofloxacin. At pH 10, ZnO nanoparticles notably demonstrated enhanced degradation of ciprofloxacin (**Table 1**).

Table 1. Photocatalytic degradation of antibiotics by pristine ZnO as photocatalyst.

Catalyst	Antibiotic	Temperature (T) °C and pH	Light Source	Degradation Efficiency (%)	Duration	Ref.
ZnO (thin film)	Paracetamol 15 ppm	T = 25 °C pH = 5.58–6.69	UV lamp (18W)	14%	4 h	[3]
ZnO powder 20 mg	Ciprofloxacin 10 mg/L	T/pH = n.a.	UV light (15 W)	100%	100 min	[3]
ZnO 1 g/L	Spiramycin 10 mg/L	T = 20 °C pH = 5.5	Xenon arc lamp (450 W)	95–99%	80 min	[4]
ZnO 25 to 75 mg	Ofloxacin (OFL) 5–20 ppm	T = n.a. pH = 10	UV light irradiation (Mercury lamp, 125 W)	95%	120 min	[4]
Nano ZnO 0.02 g/L	Ciprofloxacin 5 mg/L	T = n.a. pH = 10	Xenon lamp	50%	60 min	[5]
ZnO (thin film)	Chloramphenicol 8 ppm	T = 25 °C pH = 5.31–6.49	UV lamp (18W)	40%	4 h	[6]

Catalyst	Antibiotic	Temperature (T) °C and pH	Light Source	Degradation Efficiency (%)	Duration	Ref.
Nano ZnO 0.02 g/L	Metronidazole 10 mg/L	pH = n.a. T = 30 °C	Ultrasound irradiation	100%	27 min	[7]
ZnO nanorod array	Tetracycline hydrochloride 10 mg/mL	T/pH = n.a.	Xenon light (500 W)	69.80%	140 min	[8]

n.a., not available.

From the above findings, it can be inferred that the pH might modify the photocatalyst's surface charge features and presumably the chemical morphology of the molecule; as a corollary, the photocatalysis process is pH-dependent. It has been revealed that thin ZnO films possessed photocatalytic performance in the photocatalytic decomposition of the drugs chloramphenicol (levomycetin) and paracetamol with conditions (Table 1). When the catalyst is irradiated by ultraviolet light with either an energy equal to or higher than that of the energy band gap, an electron–hole pair is formed. Water adsorption over the ZnO layer or hydroxyl groups occurs in connection with photo-induced holes at the valence band. The outcome of such activity is the powerful OH[•] radical. Conduction band electrons react with electrophiles, such as O₂, deposited on the surface or dispersed in the water, leading to the production of super oxide radicals. By reacting with paracetamol and chloramphenicol, the ensuing strongly reactive radicals produced intermediates that degraded to give ionic species. The degradation efficiency for paracetamol and chloramphenicol under UV light was 14% and 40% with conditions (Table 1) [6]. Table 1 also summarizes numerous ZnO photocatalysts that effectively photodegraded various drug compounds, such as spiramycin (95–99%), ofloxacin (95%), metronidazole (100%), and tetracycline hydrochloride (69.8%), respectively.

2. Antibiotic Degradation Utilizing Metal-Integrated ZnO Photocatalysts

ZnO nanoparticles have several beneficial usages in piezoelectric devices, semiconductors, photovoltaic cells, polymers, skincare, and pharmaceuticals. However, they have specified drawbacks, such as: (i) potential to degrade in water solutions as a consequence of the photocorrosive effect; (ii) scattering of light, which can be minimized by modifying the catalyst dosage rate; (iii) a high energy band gap facilitates performance in ultraviolet light, which may not be realistic for massive sewage treatment; (iv) due to the recombination pathway being more rapid than the surface redox reaction, impulsive e⁻/h⁺ recombination in the photocatalyst; (v) minimal reuse. In an attempt to tackle these issues, ZnO nanomaterials have been doped with metal nanoparticles. By adding defective modes and tuning the active energy band gap, doping helps to improve the surface region of ZnO nanoparticles and also boosts their ability to absorb photons. Platinum (Pt), gold (Au), and silver (Ag) are the noble metals utilized as dopants. Noble metals easily absorb in the visible part of the electromagnetic spectrum through the surface plasmon resonance (SPR) mechanism. ZnO becomes a visible light-sensitive photocatalyst by incorporating noble metals into its crystal lattice or on its interface [9]. Transition metals, alkali metals, alkaline earth metals, as well as heavy metals such as lanthanides have also been utilized to dope ZnO photocatalysts in addition to noble metals. For instance, the minor misalignment between magnesium (Mg) and ZnO crystal structures leads to the dopant raising the energy band gap of ZnO. The basic pathway of antibiotic photodegradation by metal-doped ZnO is as follows: (i) the antibiotic drug adheres to the photocatalyst's surface; (ii) production of electron-hole pairs; and (iii) reactive oxygen species (ROS) are caused by redox processes to decompose the antibiotic [10].

Comparing the photocatalytic activity of the ZnO photocatalyst under UV light irradiation (duration: 120 min) and the Ag-ZnO photocatalyst under solar light irradiation (duration: 80 min) against the antibiotic ofloxacin revealed that 95% degradation efficiency was achieved by the ZnO photocatalyst and 100% degradation efficiency was achieved by Ag-ZnO photocatalyst (5 wt% silver) under similar operating conditions. Due to its higher electron-hole separating efficacy than pure ZnO, the Ag-doped ZnO photocatalyst exhibited complete degradation of ofloxacin. The Schottky barrier arises at the Ag/ZnO interface as metal silver is coated onto the ZnO photocatalyst, which leads to better photocatalytic activity and greater quantum efficacy [3][11]. Under ideal conditions, the Ag-ZnO photocatalyst exhibited significant degradation rates for medicinal residues, i.e., 70.2% for atenolol (ATL) and 90.8% for acetaminophen (ACT) (Table 2).

Table 2. Photocatalytic degradation of antibiotics by metal-integrated ZnO as photocatalyst.

Catalysts	Antibiotic	Temperature (T) °C and pH	Light Source	Degradation Efficiency (%)	Duration	Ref.
Ag-doped ZnO 25 mg to 75 mg	Ofloxacin 5–20 ppm	T = n.a. pH = 5–9	Solar light irradiation	100%	80 min	[3]
Mg/ZnO 1 g/L	Alprazolam 0.03 mM	T/pH = n.a.	High-pressure Hg lamp (125 W)	87%	10 min	[7]
Ag/ZnO 0.25 g/L	Tetracycline 15 mg/L	T = n.a. pH = 5–9	Visible light tungsten lamp (60 W)	100%	30 min	[8]
Ce-ZnO crystals 1 g/L	Nizatidine 5 mg/L	T = 25 °C pH = 6.7	UV-B mercury lamp (8 W)	96%	4 h	[12]
Ce-ZnO crystals 1 g/L	Levofloxacin 5 mg/L	T = 25 °C pH = 5.6	UV-B mercury lamp (8 W)	96%	4 h	[12]
Ce-ZnO crystals 1 g/L	Acetaminophen 5 mg/L	T = 25 °C pH = 6.8	UV-B mercury lamp (8 W)	65%	4h	[12]
Fe ³⁺ -doped ZnO 1 g/L	2-Chlorophenol 50 mg/L	T = 28–38 °C pH = n.a.	Solar radiation (23 W/m ²)	85%	90 min	[13]
F-ZnO 1.48 g/L	Sulfamethoxazole (SMX)	T = 21 ± 1 °C pH = 4.7	UVC lamp (10 W)	97%	30 min	[14]
La-doped ZnO	Paracetamol 100 mg/L	T/pH = n.a.	Visible light irradiation (20 W) compact fluorescent lamp	99%	3 h	[15]
Ag-ZnO NPs 1 g/L	Atenolol (ATL) 5 mg/L	T = n.a. pH = 5–9	Visible light Tungsten halogen lamp (300 W)	70.20%	120 min	[16]
Ag-ZnO NPs 1 g/L	Acetaminophen (ACT) 5 mg/L	T = n.a. pH = 5–9	Visible light Tungsten halogen lamp (300 W)	90.80%	120 min	[16]

n.a., not available.

ATL and ACT degradation is governed by pseudo-first-order kinetics. Increased surface area, improved charge transport with both ZnO and Ag, and their synergistic influence were the main factors that contributed to the increase in efficiency. An additional finding about the photocatalyst mechanism revealed that the main method for removing ATL and ACT was the OH[•] pathway [4]. The Ag/ZnO synthesized using the rapid, one-pot, surfactant-free, microwave-assisted, aqueous solution method showed complete degradation of the antibiotic tetracycline (TC) in 30 min under visible light [7]. Silver nanoparticles act as an electron sink in Ag-doped ZnO nanoparticles, improving the charge separation. This enhances the generation of hydroxyl radicals in the reaction media and consequently improves the photocatalytic performance of ZnO nanoparticles [17]. Employing a conventional solid-state process, Mg-doped ZnO nanocrystallites were synthesized and exhibited an 87% alprazolam degradation rate. Mg-doped ZnO annealed at 700 °C showed excellent photocatalytic properties, reaching 100% degradation within 20 min. Meanwhile, under similar conditions, bare ZnO showed 78% removal of alprazolam. Due to excellent electron-hole separation and good textural characteristics, doping ZnO nanoparticles with Mg enhanced the photocatalytic performance triggered by sunlight and the results indicated that doping ZnO with Mg promoted alprazolam elimination by 10% [6]. Under UV light, a hydrothermally prepared Ce-doped ZnO photocatalyst demonstrated 96% degradation rates for nizatidine and levofloxacin and a 65% degradation rate for acetaminophen [12]. Research revealed that the metal doping of ZnO was necessary for controlling the ZnO nanoparticle surface and optical features. It must be highlighted that the photodegradation of metal-doped ZnO depends on the light source. This is due to ZnO's ability to absorb UV light whereas metal-doped ZnO absorbs light primarily in the visible spectrum. Therefore, due to the fact that sunlight carries UV along with visible light, the excitonic formation under solar irradiation may arise by either ZnO or the doped metal [17].

3. Antibiotic Degradation Utilizing ZnO Composites Photocatalysts

The major concern with ZnO in photocatalysis is mainly its high rate of electron-hole recombination. With the aim of enhancing the performance of photocatalysis, hetero-junction semiconductors have been developed and used to prevent

electron-hole recombination [18], consequently enhancing the degradation efficiency with better regeneration and recycling.

The photodegradation of tetracycline by the ZnO/ γ -Fe₂O₃ composite showed 88.52% degradation efficiency under UV-visible light over 150 min. Both the catalyst's surface and pore volume were increased by the inclusion of iron oxide in its structure, thereby promoting the analyte's adsorption on the nanostructure's surface, which is a crucial step in improving photocatalytic degradation. Additionally, it was observed that ZnO served a vital function in the photocatalytic degradation supported by γ -Fe₂O₃, raising the rate of TC degradation to 20% [16]. The photodegradation activity of the ZnO globular-gC₃N₄ nanocomposite showed that 78.4% of tetracycline degraded in 50 min and 63.5% of oxytetracycline degraded in 50 min. Reactive species were regarded as the primary cause in the photodegradation mechanism [13]. As Ag loading rose during the photocatalytic degradation of the antibiotic tetracycline hydrochloride, the catalytic performance of the Ag@ZnO/BiOCl composite first rose and then fell. ZnO has minimal photocatalytic performance and achieved a degradation rate of 38.5% over 80 min. The degradation rate achieved by ZnO/BiOCl was moderately greater, attaining 42.7%. It is noteworthy that the degradation rates of Ag-loaded samples were better than that of ZnO/BiOCl, and the Ag nanocomposite achieved the maximum degradation rate with 80.4% removal of tetracycline hydrochloride (Table 3).

Table 3. Photocatalytic degradation of antibiotics by ZnO nanocomposites as photocatalysts.

Catalyst	Antibiotic	Temperature (T) °C and pH	Light Source	Degradation Efficiency (%)	Duration (min)	Ref.
ZnO/g-C ₃ N ₄ 20 mg	Tetracycline (TC) 20 mg/L	T/pH = n.a.	Visible light (300 W)	78.40%	50	[15]
ZnO/g-C ₃ N ₄ 20 mg	Oxytetracycline 20 mg/L	T/pH = n.a.	Visible light	63.50%	50	[15]
ZnO/ γ -Fe ₂ O ₃ composite 0.5 mg/mL	Tetracycline (TC) 30 mg/L	T = RT pH = 6.7	UV-visible light (100 mW cm ⁻²)	88.52%	150	[16]
Ag Loaded ZnO/BiOCl 32 mg	Tetracycline hydrochloride 20 mg/L	T = 25 °C pH = 8	Simulated solar light	80.40%	80	[19]
SnO ₂ /ZnO nanocomposite 50 mg	Ciprofloxacin 20 mg/L	T/pH = n.a.	Mercury lamp (300 W)	91.23%	60	[20]
SnO ₂ /ZnO nanocomposite 50 mg	Ofloxacin 20 mg/L	T/pH = n.a.	Mercury lamp (300 W)	91.26%	60	[21]
SnO ₂ /ZnO nanocomposite 50 mg	Norfloxacin 20 mg/L	T/pH = n.a.	Mercury lamp (300 W)	88.39%	60	[22]
ZnO/GO/DES nanocomposite 0.532 g/L	Cefxime trihydrate 20.13 mg/L	T = 40.0 ± 1 °C pH = 4.03	UV-A irradiation (15 W)	86%	60	[23]
GO@Fe ₃ O ₄ / ZnO/SnO ₂ composite 1 g/L	Azithromycin 30 mg/L	T = n.a. pH = 3	UV-C irradiation (6 W)	90.06%	120	[24]
Ag-ZnO/GP composite 0.5 g/L	Metronidazole (MNZ) 30 mg/L	T = n.a. pH = 9	UV-lamp (100 W)	88.50%	60	[25]
Ag-ZnO/GP composite 0.5 g/L	Metronidazole (MNZ) 30 mg/L	T = n.a. pH = 9	Solar irradiation (500 W)	97.30%	180	[25]

n.a., not available.

This was achieved as a result of enhanced absorption of visible light, better charge separation as a function of surface plasmon resonance, the potential of Ag to capture electrons, and enhanced surface catalysis. Hence, adding noble metals to a semiconductor with a wide energy band gap is a practical approach to improving its photocatalytic performance [19]. A simple hydrothermal method was utilized to synthesize the SnO₂/ZnO nanocomposite investigated for the photocatalytic degradation of quinolone antibiotics (ciprofloxacin, ofloxacin, and norfloxacin). Over 60 min, ciprofloxacin, ofloxacin, and

norfloxacin were all degraded at rates of 91.23%, 91.26%, and 88.39%, respectively. Strong oxidation potential is present in SnO₂, whereas high reduction ability is found in ZnO. This can substantially increase the separation efficacy of photo-induced e⁻/h⁺. Therefore, a simple method for improving the photocatalytic properties is to combine SnO₂ with ZnO to produce a composite photocatalyst [20]. The photodegradation of the most used antibiotics by several ZnO nanocomposites is summarized in **Table 3**. Ag–ZnO/GP composite, SnO₂/ZnO nanocomposite, GO@Fe₃O₄/ZnO/SnO₂ composite, and ZnO/γ-Fe₂O₃ composite are recognized for exhibiting the maximum degradation for the frequently prescribed antibiotics metronidazole, ciprofloxacin and ofloxacin, azithromycin, and tetracycline, respectively. Therefore, ZnO nanocomposites are potential and effective preferences for the photocatalytic degradation of antibiotics.

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