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ORIGINAL ARTICLE

Improved remediation of contaminated water using ZnO systems via chemical treatment: applications, implications and toxicological mitigation

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Abstract

Environmental remediation is an alternative field of science that can solve various environmental challenges based on numerous treatment methods. In particular, heterogeneous photocatalysis is an advanced oxidative process that has been the subject of many studies related to environmental control. Globally, pollution by organic pollutants represents risks to environmental health that compromise public health and directly affect the scientific knowledge of public policies that potentially improve quality of life from a sustainable point of view. ZnO systems are receiving special attention due to their attractive characteristics (non-toxic nature, high surface area, thermal/chemical stability), availability of being chemically modified by various strategies, and particularly good environmental remediation. This review focuses on the efforts of ZnO-based photocatalysts, such as the methods, chemical modifications, operational parameters, and the effects after the release of effluents in aquatic matrices. Therefore, we investigated the recent advances in zinc systems aimed at treating contaminated water and their direct application in environmental remediation.

Keywords Environmental remediation · Zinc oxide · Photocatalytic process

Introduction

The increase in water consumption and the correspondingly high levels of pollution have generated a prominent need for water management and quality (Speight 2020). Urban rivers, lakes, and streams are being gradually affected and reduced in many countries and regions due to scaled discharge of industrial and agricultural effluents, mining tailings, which includes; mining sewer systems, and wastewater treatment plants, emerging contaminants, and toxic metals, which causing negative impacts on the ecosystem (Jiang 2009; Sabater et al 2018).

Annually, thousands of different molecules with toxic potential are released into the environment, generating adverse effects on aquatic biota, food safety, water quality, and environmental sustainability (Riva et al. 2019). Emerging contaminants, for example, are chemicals of natural or synthetic origin that need to be monitored and regulated in the environment (Vashisht et al. 2020). These contaminants include; pharmaceuticals, pesticides, metals, surfactants, industrial effluents, solvents, plasticizers, and hormones. The threat of these contaminants and their complex mixtures is concentrated in the occurrence, destination, transport, toxicity, and recovery. It lies in the risks and undesirable effects for aquatic and terrestrial organisms, potentially, humans given the high biological activity, given the high biological activity and the significant deficit of treatment (Brausch and

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Rand 2011; Ternes et al. 2015), and the difficulty of recovery due to the nature and degree of risk posed by the contamination (Vashisht et al. 2020). Furthermore, when these contaminants are subjected to cleaning treatment, by-products are generated whose chemical properties are undetermined (Vashisht et al. 2020).

The Handbook of Environmental Chemistry cited by Rosenfeld and Feng (2011) emphasizes the danger of these compounds by stating the following point. Every day new emerging contaminant is discovered, and new disinfection by-products are also generated during treatment, with complete ignorance of their toxicity with potential or effect on human health (Rosenfeld and Feng 2011), sustainable development goals (SDGs) (Ghiasi and Malekzadeh 2014; Yusuff et al. 2019; Rai 2022). Several techniques for the remediation of contaminated areas are used to achieve the established goals. Among the main advanced remediation techniques is the remediation of contaminated areas by multiphase extraction (MPE), by chemical oxidation in situ, by removal of contaminated soil, by thermal remediation, addition to physical, chemical, electrochemical and biological processes discussed and developed around the world (Silva et al. 2018; Yusuff et al. 2019; Wu et al. 2020). Advanced Oxidative Processes (AOP) requirements for the purification and cleaning of contaminated waters due to their high performance and simplicity (Lado Ribeiro et al. 2019; Eshaq et al. 2020). It is advantageous over other competing methods as it provides complete mineralization, does not result in solid waste disposal problems, and does not generate thermal pollution (Yusuff et al. 2019).

Heterogeneous photocatalysis has gained prominence as an oxidation technique among the most popular photochemical treatment methods. Due to its heterogeneous nature, that allows it to treat water and industrial effluents in a wide pH range (Chen et al. 2020; Lum et al. 2020; Ahmad et al. 2020b), through reactions usually regulated by a free radical mechanism ($OH^{\bullet}$, $O_2^{\bullet -}$, $HO_2^{\bullet}$)(Colmenares et al. 2017). The main attributes of this technology include the ability to use solar and/or artificial energy, low temperature and pressure conditions, low process cost, energy feasibility, susceptibility, and development of environmentally friendly and economically viable technologies with low production of tailings (Honorio et al. 2019; Mohamed Isa et al. 2021).

The demand for efficient photocatalysts is growing due to their ease and cost-effectiveness. The zinc oxide (ZnO) has been a constant focus of research and reviews in the photocatalytic area, being a promising photocatalyst with high visibility in the photochemical industry (Sanzone et al. 2018; Ruiz-Hitzky et al. 2019). By comparing important aspects, such as low cost, low toxicity, and optical properties, ZnO has contributed to advances in this segment (Mohamed et al. 2020). Based on this context, the following sections will describe the fundamentals of zinc oxide nanoparticles (pure, doped, heterostructured, immobilized, and supported) applied in degradation processes and their operational conditions for the oxidation of substances defined as organic contaminants, for example, synthetic dyes (Ahmad et al. 2021; Freitas et al. 2022), drugs (Mohamed Isa et al. 2021; Nasseh et al. 2022), and pesticides (Behera et al. 2021; Ahmad et al. 2022).

Fundamentals of heterogeneous photocatalysis

Photocatalysis has attracted great attention in many research fields due to its potential to combat environmental impacts (Chen et al. 2020). Historically, photocatalysis has been the principle of photochemistry with reports since 1911, using titanium oxide (TiO_2) as a semiconductor for the first time (Serpone et al. 2012; Ahmad et al. 2020b). Its epistemological origin is attributed to the combination of "*fos* and *katalyo*" from the Greek family, which means light and decompose/degrade, respectively (Lum et al,. 2020; Ahmad et al. 2020b). In 1972, Fujishima and Honda described the process of separating water using TiO_2 electrodes under UV irradiation, generating hydrogen and oxygen (Fujishima and Honda 1972; Saravanan et al. 2017). Since then, several studies have explored the photoactive properties of semiconductors, especially TiO_2. Focusing mostly on pollutant degradation, water purification, and H_2 production obtained through water splitting (Ahmad et al. 2020b; Rodríguez-González et al. 2020), the number of publications in the field of photocatalysis has increased annually. Figure 1 charts the linear profile

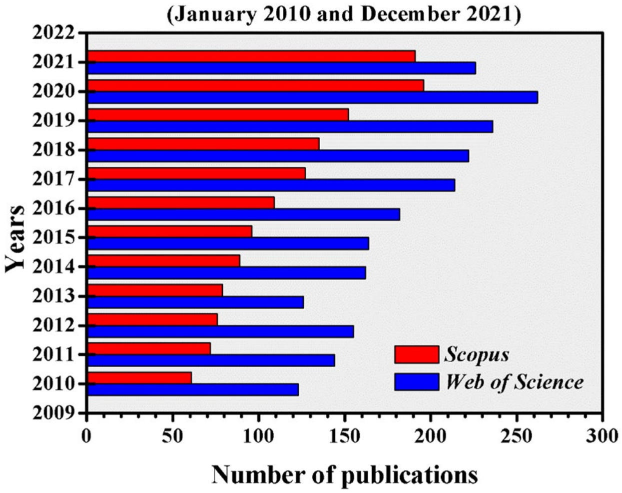

Fig. 1 Annual representation of the number of articles published and identified in the Web of Science and Scopus databases related to the term "heterogeneous photocatalysis" between January 2010 and September 2020

of the use of this term (heterogeneous photocatalysis) as a technology for environmental management over the last decade in the Web of Science and Scopus database.

The main attributes of this technology basically include the ability to use solar and/or artificial energy, low temperature and pressure conditions, low process cost, energy feasibility, susceptibility, and development of environmentally friendly and economically viable technologies with low production of tailings. The feasibility of using photochemical technologies is emphasized by the possibility of reusing photocatalysts (operational recycling) and by the high reactive yield. It often allows the complete destruction of target pollutants of different chemical classes, forming residues with less environmental impact (Cavalcanti et al. 2019; Khan and Narula 2019; Wetchakun et al. 2019; Liu et al. 2020), thus encompassing the concept of "green treatment"(Honorio et al. 2019; Mohamed Isa et al. 2021).

A photocatalysis is defined as a technology that involves a photoinduced reaction accelerated by the presence of a semiconductor bombarded by photons of a natural or artificial nature (Kabra et al. 2004; Qian et al. 2019). In addition, its reactions and mechanisms vary and can be described in stages (Zhu and Wang 2017; Al-Mamun et al. 2019). Its general concept is understood by mechanisms of excitations and generations of hydroxyl radicals (Al-Mamun et al. 2019). The fundamental characteristic of semiconductors is the energy discontinuity between the valence band (VB—lowest energy region) and the conduction band (CB—highest energy region), with the difference being called "band gap" (Wei et al. 2016; Saravanan et al. 2017). When the absorption of a photon (*hυ*) is equal to or greater than the energy of the band gap, electrons (e^-) are promoted from VB to CB, leaving a positive hole (h^+) in VB. Thus creating an electron/hole pair, (e_{CB}^-/h_{VB}^+), or exciton(Byrne et al. 2018; Hasanpour and Hatami 2020), whose schematic representation of the mechanism is displayed in Fig. 2.

The reductive-oxidative capacity of electrons and photogenerated holes is determined by the conduction band and the potential of the valence band of the respective semiconductor particle (Molinari et al. 2017; Saravanan et al. 2017). Oxidation reactions can occur through the VB hole and H_2O, producing hydroxyl radicals (Teixeira and Jardim 2004). Reduction reactions may occur between the CB electron and molecular oxygen, producing the superoxide ion. This free radical can decompose and produce hydrogen peroxide, which in turn produces hydroxyl radicals (Teixeira and Jardim 2004; Saravanan et al. 2017).

According to Qian et al. (2019), photocatalytic efficiency is comprised of two parts. The competitiveness between charge carrier recombination and entrapment (part one) and the interfacial charge transfer (part two). Both are important to understand the degradation kinetics and therefore the photocatalytic mechanism. Such mechanisms are reported in the literature and classified as direct and indirect (Rauf and Ashraf 2009; Rauf et al. 2011).

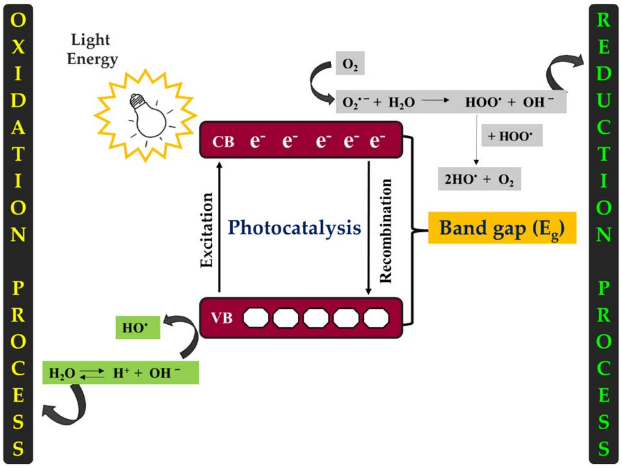

Fig. 2 Simplification of the photocatalytic mechanism using a semiconductor

The photoactivity of a semiconductor in a photocatalytic process will depend greatly on characteristics. Such as the composition of crystals (crystalline structure), surface area, particle size distribution (adequate morphology), porosity, hydroxyl density on the surface, and adsorption characteristics/desorption, with band gap energy being an important property in the photogeneration of electron–hole pairs in semiconductors (Hisatomi et al. 2014; Qayyum et al. 2018).

Zinc dioxide (ZnO) has been a constant focus of research and reviews in the photocatalytic area. It is a promising photocatalyst with high visibility in the photochemical industry (Sanzone et al. 2018; Ruiz-Hitzky et al. 2019). By comparing important aspects, such as low cost, low toxicity, and optical properties, ZnO has contributed to advances in this segment (Qi et al. 2017; Vishnukumar et al. 2018; Mohamed et al. 2020). Under UV irradiation, both TiO_2 and ZnO are highly efficient photocatalysts, since their electrons and photogenerated holes are oxidizing and reducing species, respectively (Janotti and Van de Walle 2009; Kavitha and Kumar 2019).

Strategies to improve the photocatalytic activity of ZnO

ZnO is one of the main photocatalysts applied in the management of environmental waste due to its optoelectronic, piezoelectric, catalytic, and photochemical properties (Zheng et al. 2019; Le et al. 2020), and its high quantum efficiency, which provides a significant advantage. It is an n-type semiconductor with a band gap around 3.37 eV (UV region) and a considerable free-exciton binding energy (so that excitonic emission processes can persist at or even above room temperature) (Hanh et al. 2019; Le et al. 2020). ZnO's most common crystalline phase is the wurtzite (hexagonal) structure at room temperature. In this structure, the Zn^{2+} cation is surrounded by four O^{2-}anions (Lee et al. 2016; Le et al. 2020). This tetrahedral atomic arrangement causes a non-centrosymmetric reaction in the ZnO crystal structure (Le et al. 2020). One of the relevant characteristics of ZnO is the performance of optical devices with piezoelectric properties covering applications in the field of energy technology, used in the production of closed diodes, laser diodes and photodetectors, field-effect transistors, pressure-based sensors, gas sensors, biomedical sensors, solar cells, gas detection, photovoltaic devices, and photocatalyst (Hou and Liu 2020; Bhati et al. 2020; Pinho et al. 2020). Figure 3 illustrates some functions and applications of ZnO.

From a photocatalytic point of view, the literature has addressed several strategies (doping of metal ions, doping of non-metallic immobilization, deposition, incorporation, heterojunctions, surface sensitization) to extend its light absorption to the visible range, modifying the surface of the compounds and improving photocatalytic efficiency (Abebe et al. 2020; Ahmad et al. 2020a; Ates et al. 2020; Peng et al. 2021; Sá et al. 2021). In addition, such modifications improve performance by changing the stability of particles to create structures that function as excitonic traps and regions with different redox potentials (Zhu et al. 2020).

Velmurugan et al. reported that ZnO nanoparticles from zinc oxalate dihydrate are more efficient than commercial ZnO for degrading reactive red 120 (RR120) under sunlight. Those authors attributed the efficiency of ZnO nanocrystals to the transfer of electrons from the sunlight-sensitized dye molecule to the nano-ZnO CB, which promoted an increase in the generation of free radicals and consequent degradation of the dye (Velmurugan and Swaminathan 2011).

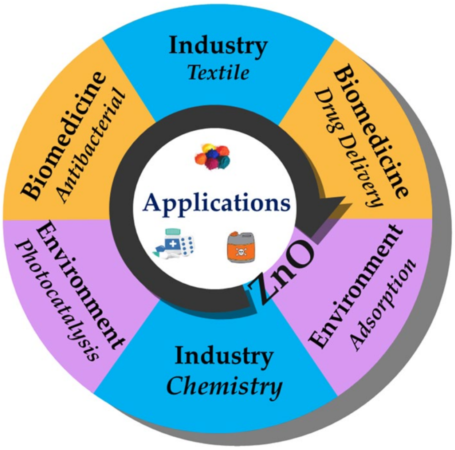

Fig. 3 Various applications of ZnO in the field of biomedicine, industry and environment

Fe_2O_3 nanoparticles were selected as a sample to manufacture the multi-composite photocatalyst to improve the photocatalytic activity of ZnO and TiO_2 (TiO_2/Fe_2O_3 and ZnO/Fe_2O_3 based on clinoptilolite) to extend its band gap to the visible region (Davari et al. 2017). Davari et al. synthesized and evaluated the operational parameters in the degradation of the drug diphenhydramine (DPH). In the photocatalytic evaluation, ZnO/Fe_2O_3/Zeolite exhibited a more effective performance for the degradation of DPH than TiO_2/Fe_2O_3/Zeolite. The photocatalytic degradation of DPH with ZnO/Fe_2O_3/Zeolite reached 95% at optimal conditions (Davari et al. 2017).

ZnO nanoparticles doped with lanthanum were used in the photocatalytic degradation of paracetamol. The doping process is essential in the optical absorption of visible light due to reducing particle size and gap energy. The doped photocatalyst showed more excellent photocatalytic activity than pure ZnO, reaching 99% of paracetamol degradation after three hours of irradiation (Thi and Lee 2017). Furthermore, the HPLC and GC–MS analyses demonstrated that the detected photocatalytic products aided the proposed mechanism, including the generation of radicals and subsequent reaction pathways (Thi and Lee 2017).

Sanad et al. prepared ZnO, ZnS, and ZnO@ZnS nanocomposites by the sol–gel method. They evaluated the degradation of methylene blue and eosin dyes based on the use of scavengers of active species to understand the pathways in degradation. The incorporation of ZnO onto the surface of ZnS alters the stabilization of the oxide particles, reducing the effect of recombination between the charges (e^-/h^+) and favoring the destruction of dyes (Sanad et al. 2018).

Bozkurt Çırak et al. manufactured ZnO nanocomposite syntheses decorated on TiO2 nanotubes using two stages. In addition, different ZnO deposition cycles were used to degrade rhodamine B (RhB) dye under UV radiation. The decorated films exhibited improved photocatalytic activity (UVC, UVB, UVA, and visible region after 10 minutes were 100, 44, 22, and 4%, respectively) and good photostability after four consecutive degradation cycles (Bozkurt Çırak et al. 2019).

Abukhadra et al. (2020) synthesized kaolinite nanotubes anchored by ZnO nanoparticles (ZnO/KNTs) and evaluated the photocatalytic oxidation of the antibiotic levofloxacin under a visible light source. ZnO/KNTs showed excellent results of 99.3, 99.6, and 99.8% by varying the dosages of photocatalyst and pH of the reaction medium. In addition, degradation was confirmed by the detection of residual TOC after analysis of the tests, and the photogenerated intermediates served to elucidate the steps of the mechanism since hydroxyl radicals were the main oxidizing species of the system (Abukhadra et al. 2020).

The g-C_3N_4/ZnO nanorods were prepared by simple hydrothermal, grinding, and calcination methods. The nanobasts were evaluated to degrade several essential pollutants such as MB, RhB, Cr (VI), and eosin (Zhong et al. 2020). The improved photocatalytic performance of photocatalytic nano bonds maintained high stability after five degradation cycles reaching 97% degradation (for MB) and consequently the great potential for the treatment of water pollutants. Furthermore, the tests of radical scavengers and ESR proved the degradation by proposing a photocatalytic mechanism that indicates the $O_2^{\bullet-}$ and $^{\bullet}OH$ species as majorly significant in the degradation process (Zhong et al. 2020).

Among the methods of modifying ZnO, doping aims to improve photocatalytic performance by replacing and/or introducing a doping agent into the matrix network (Hanh et al. 2019; Shah et al. 2020). The doping effect, described by Reddy et al. (Neelakanta Reddy et al. 2018), can improve the electrical and optical properties of the semiconductor through the accumulation of impurities or the construction of intrinsic defects. In addition, doping involves forming new electronic levels inside the semiconductor, facilitating the narrowing of the bandgap and contributing to the separation of charges from the original material (Neelakanta Reddy et al. 2018). Several other dopants, including Fe, Ag, Ni, Co, Mn, Ce, and Cu (Kharatzadeh et al. 2021; Yang et al. 2021), are used to enhance the photocatalytic activity of ZnO nanoparticles (Al Abri et al. 2019; Jyothi and Ravichandran 2020).

The complexity of producing the desired property is due to phase purity, crystallinity, stoichiometry, size, and morphology (Wojnarowicz et al. 2020). Many conventional routes for preparing ZnO-based nanostructures employ physico-chemical methods (co-precipitation, precipitation, hydrothermal, thermal decomposition, microemulsion, sonochemical, ultrasonic, solid-state, sol–gel) that require specific configuration, high cost, high temperature–pressure conditions, and non-ecological chemicals (Akhtar et al. 2020; Gu et al. 2020; Wojnarowicz et al. 2020; Weldegebrieal 2020; Masmali et al. 2021). Fig. 4 presents synthesis methods used in the preparation of ZnO-based photocatalysts according to the literature (Shirdel and Behnajady 2020; Tsai et al. 2021; Kampalapura Swamy et al. 2021; Bharathi et al. 2022; Dineshbabu et al. 2022).

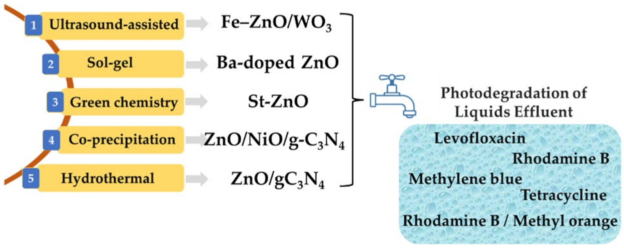

Fig. 4 Various synthesis methods used in ZnO photocatalysts according to the literature

Another method that has been widely used is the green synthesis approach. Due to the elimination of chemical products of toxic nature and the application of environmentally correct and viable routes since it eliminates the use of toxic chemicals and applies ecological routes (Isık et al. 2019; Weldegebrieal 2020). Weldegebrieal synthesized biomolecules based on ZnO using plant extracts through the biosynthesis method. They pointed out that both the bactericidal efficacy of the compounds and the photodegradation efficiency is strongly affected by several factors such as the size, shape of the particles and the method of synthesis employed (Weldegebrieal 2020).

Parameters affecting the photoactivity of ZnO

Photocatalysts are materials whose VB and CB are separated by an energy gap widely used in photoinduced processes (Araujo et al. 2020a; Wang et al. 2020; de Sousa Filho et al. 2020; Miranda et al. 2020). Since the texture, size, and surface characteristics significantly influence the photocatalytic efficiency of metals and semiconductors, as well as their electronic and optical properties, synthetic methods are expected to lead to reliable results that obtain particles with low polydispersity, uniform size, and morphology defined in dimensions 1D, 2D, and 3D (Radhika et al. 2019). Photocatalytic degradation performance is also closely related to operational conditions that vary according to established physical–chemical tests and parameters. Such parameters that positively and/or negatively affect the process can also be optimized to ensure more excellent catalytic conversion and lower economic costs (Malato et al. 2009; Reza et al. 2017; Anwer et al. 2019; Faisal et al. 2019).

The effect of the initial concentration of the contaminant

The primary mechanism of the photocatalytic process involves the absorption of light and the formation of radical species that attack the contaminant. Therefore, the greater the concentration of contaminants, the higher the degradation rate (which is expected from the kinetic point of view), taking into account the adsorption phenomena that make the degradation more effective. After reaching the critical concentration value, which is the maximum value supported by the charge of the photocatalyst, the degradation rate is reduced due to the many layers by the adsorption of contaminant on the catalyst surface. This blocking layer makes it impossible to produce new radicals. Strategies are needed significantly to modify the photocatalyst surface structures, to avoid or minimize this negative effect caused by the high concentration of pollutants (Teixeira and Jardim 2004; Chakraborty et al. 2017; Honorio et al. 2019).

The effect of the amount of catalyst

The general trend is that greater concentration increases activity due to the more excellent radiation absorption, thus creating radicals (Ebrahiem et al. 2017; Chen et al. 2020). At high catalyst concentrations, the aggregation of the particles with a decrease in the effective surface is a limiting factor due to light scattering with a decrease in UV light penetration (Chen et al. 2020). Light scattering phenomena caused by the accumulation of suspended particles hinder light infiltration and the production of radical species (Saravanan et al. 2017). Wang and coworkers (2007) found that with the considerable increase in the amount of catalyst, the surface that adsorbs the photon is not increasing in a geometrical ratio due to inclination towards aggregation (Wang et al. 2007). Therefore, the quantity in mass ratio, or the fraction and mass, between the photocatalyst and the contaminant is significant to achieve the best photocatalytic conversion rates (Rodrigues et al. 2008; Çalışkan et al. 2017).

The effect of pH

Understanding the real influence of pH is complex due to its multiple functions (Akpan and Hameed 2009). For example, changes in pH can influence the adsorption of organic molecules by modifying the surface charge of photocatalysts (Ajmal et al. 2014; Reza et al. 2017) and the adsorption of contaminants being controlled or dominated by electrostatic forces.

Zhu et al. proved that the effect of pH is directly related to the surface charge properties of the photocatalysts and can be explained based on the point of zero charges (PZC) (Zhu et al. 2012). For ZnO pHpzc is in the pH range 8–9 (Sukriti et al. 2020). At pH values less than pHpzc or in an acidic solution, the surface of the photocatalyst is positively charged, whereas it is negatively charged under alkaline conditions (i.e.pH > pHpzc) (Akpan and Hameed 2009; Anwer et al. 2019).

In the case of dye degradation, its nature can undergo protonation or deprotonation and change its adsorption characteristics and redox activity (Konstantinou and Albanis 2004). Moreover, high pH often causes high hydroxylation of the catalyst surface due to a large number of $OH^{\cdot}$ ions. Therefore, it facilitates the photogeneration of more hydroxyl radicals and consequently enhances the photocatalytic degradation efficiency (Caregnato et al. 2021). The effect of pH on the photocatalytic degradation of contaminants has been extensively investigated (Chen et al. 2017).

The nature and intensity of light

The operational principle of the photocatalytic reaction, the nature of light, or better, the electromagnetic radiation that provides energy to activate the photochemical mechanism, is one of the essential parameters of the phenomenon. The effective absorption of light by the photocatalyst will depend mainly on the distribution of wavelengths and the power of the light source. These characteristics directly affect the formation process of oxidative species and other chemical/physical factors hindering and/or favoring degradation (Colmenares et al. 2017; Sergejevs et al. 2019).

The effect of oxidizing agent

The action of oxidizing agents influences the photocatalytic degradation of pollutants such as dye, hydrogen peroxide (H_2O_2), ammonium persulfate ($(NH_4)_2S_2O_8$), and potassium bromate ($KBrO_3$) (Akpan and Hameed 2009). These oxidants can trap electrons, preventing recombination, which generates radical oxidation and consequently increases photocatalytic degradation (Reza et al. 2017). Among oxidizing species, H_2O_2 is widely studied in advanced oxidation processes in wastewater treatment (Sakarkar et al. 2020). H_2O_2 plays a dual role during the degradation process, as it acts by accepting electrons from CB (Conduct Band), promoting the separation of charge carriers, and forming radical OH (Ajmal et al. 2014).

Whether through hydroxyl radicals or direct oxidation through holes, the preferred mechanism of photooxidation remains a matter of debate. The critical point is that many studies evaluate not only the efficiency of the reaction process but also the influence of active species based on the use of inhibitors and/or scavengers of electrons and holes. It indicates a possible orientation and the importance of each species in the targeted reaction in suppressing the recombination of charges (Gusmão et al. 2019; Araujo et al. 2020b; Honorio et al. 2020; Freitas et al. 2022).

The choice of organic contaminants in the photocalytic reaction

The choice of pollutants is the main factor in knowing the photocatalytic materials, mainly when synthesized. This section will describe which are the most used and how materials can influence the presence of these molecules. Water pollution poses a significant threat to the survival of modern society (Rasheed et al. 2020). The thousands of water pollutants include dyes, pesticides, detergents, drugs, heavy metals, radioactive substances, polyaromatic hydrocarbons, plasticizers, phenolic compounds, and industrial solvents, categorically divided into different classes and compositions (Rasheed et al. 2020). In addition, organic pollutants, also known as emerging pollutants (EPs), are chemical compounds with a natural or synthetic origin, which are not commonly monitored in water, soil, and air but can cause severe problems to the environment and human beings (Geissen et al. 2015). Examples of emerging pollutants include drugs (ibuprofen, azithromycin, diclofenac, losartan, and ciprofloxacin), dyes (methylene blue, curcumin, lycopene, and rhodamine B), pesticides (glyphosate, dichlorophenoxyacetic acid, and atrazine Tkaczyk et al. 2020; Sun et al. 2020).

Dyes

Dyes are organic compounds that strongly absorb wavelengths in the region of visible light, which is why they are colored (Brillas and Martínez-Huitle 2015). Their molecules contain two main groups: the chromophores responsible for the color and the auxochromes that determine the intensity of the color (Arora 2014). Classification of dyes is based on the chemical class to which they belong and the applications to which they are intended (Guaratini and Zanoni 2000; Honorio et al. 2019; Benkhaya et al. 2020a; Samsami et al. 2020), as shown in Fig. 5.

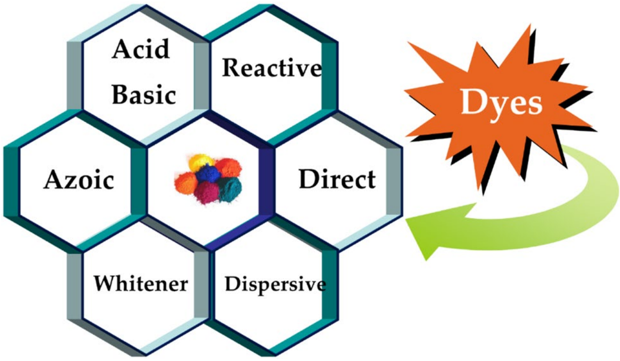

Fig. 5 Classification of dyes based on chemical class found in pollutant degradation

Ajmal et al. and Guaratini et al. current definitions and characteristics of the dyes. Direct dyes are designated as having interactions with the fibers through electrostatic forces; acid dyes are generally applied to nitrogen fibers or tissues in solutions of organic or inorganic acid; and dispersed dyes present low solubility in water. (Ajmal et al. 2014) (Guaratini and Zanoni 2000). However, they can interact with the polyester chains forming dispersed particles, among other corresponding characteristics.

Among the main categories of dyes used in industries, the azo type has the most significant chemical production volume, playing an essential role in dyeing, especially printing production (Benkhaya et al. 2020b). Azo dyes are characterized by the group –N = N– linked to aromatic groups, benzene rings, naphthalenes, aromatic heterocyclics, or aliphatic groups and constitute 70% of the total production used in the industries(Rauf et al. 2011; Sen et al. 2016). These dyes can behave as anionic (deprotonation in the acid group), cationic (protonated in the amino group), or non-ionic, depending on the pH of the medium (Ajmal et al. 2014). Due to inefficiency during the dyeing process, they accumulate in water bodies and cause adverse effects related to dissolved oxygen (DO), biological oxygen demand (BOD), and chemical oxygen demand (COD), in addition to being highly resistant to chemical and photochemical degradation processes. This problem urgently requires new treatment routes to eliminate and/or convert them into functional and safe products (Teixeira and Jardim 2004; Benkhaya et al. 2020b).

Among the azo class, one of the most used dyes as a model pollutant is the methylene blue (MB) dye, also known as tetramethylthionine chloride, is a cationic dye that has a heterocyclic aromatic structure and is widely used as a model for the removal of dyes from aqueous solutions (Neris et al. 2019; Jaleh et al. 2021). Widely used in the textile industry, when discarded without prior and appropriate treatment, MB poses severe risks for contamination of the environment and ecosystems, intoxication, and to human beings (Phuruangrat et al. 2019; Hernández-Carrillo et al. 2020).

In 2019, ZnO and Ag-ZnO (doped) nanocomposites synthesized via ultrasound were investigated considering the degradation of methylene blue (MB) dye in an aqueous solution under sunlight irradiation (Satdeve et al. 2019). Several operational parameters were discussed, such as the effect of the initial dye concentration, catalyst dosage, and pH of the solution to observe the experiments optimization. The maximum activity reached 96.2% at the end of 120 min, confirming that ZnO doped with Ag presents better degradation concerning pure ZnO (89.77%) under similar conditions. The high activity is explained by the decrease in the recombination rate of the electron–hole pair with efficient transfer of electrons to dissolved oxygen, leading to the formation of radical species. In addition, a comparative study on MB degradation using the Ag-ZnO nanocomposite synthesized by different synthesis methods was observed, showing that the Ag-ZnO nanocomposite ultrasonically showed better degradation of the MB dye. Due to the fine dispersion of Ag in ZnO and the particle size that favored the increase in the surface area, more significant degradation of the MB dye (Satdeve et al. 2019).

Ramos-Corona et al. (Ramos-Corona et al. 2019) evaluated a new system based on ZnO doped with nitrogen (ZT) and supported on graphene oxide (GO) in the degradation of the methylene blue dye. Compared to the ZnO and ZT photocatalysts, ZTGO reached 95% degradation of the MB under UV energy in 35 min. The activity was attributed to the GO, which has a synergic effect with the N-doped ZnO as a collector and electron transporter. (Ramos-Corona et al. 2019).

In 2020, ZnO structures with flower-shaped morphology were obtained in conjunction with AG (Arabic Gum) and KG (Karaya Gum) polysaccharides, and the formation of the hexagonal structure of ZnO present in the structure was confirmed (Araujo et al. 2020c). Due to the better optical and morphological properties, the photocatalyst KGZnO showed better performance in the photodegradation of the pollutant under irradiation of visible light. Photophysical processes and photocatalytic activity were essential in interpreting the behavior between the dynamic separation of charge carriers and the recombination process (Araujo et al. 2020c). The presence of hijackers of active species and the study of toxicological assessment using *artemia salina* as a bioindicator were investigated (Araujo et al. 2020c).

Pharmaceutical drugs and pesticides

Although advances in the pharmaceutical industry are extremely relevant and beneficial to human and animal health, significant amounts of pharmaceutical products are released into the environment causing risks and uncertainties associated with a lack of knowledge about their fate, absorption, metabolism, excretion, incorrect dumping, and persistence. They are the main forms of contamination that impact the lives of people, the environment, and ecosystems (Mackuľak et al. 2017; Rastogi et al. 2018). Pharmaceutical products are considered emerging contaminants due to their increasing presence in environmental systems, such as surface water (rivers, oceans, and lakes), groundwater, and potable water (after conventional water treatment). Their high biological activity, limited information on their effects on aquatic ecosystems, and formation of metabolites of unknown nature (Bade et al. 2018; Castrignanò et al. 2018). In addition, to being substances absorbed by the body, cause uncertainty about their effects (Bila and Dezotti 2003).

Ebele et al. define pharmaceutical products as over-the-counter and prescription medications used for therapeutic and veterinary uses with the purpose of preventing and/or treating diseases and guaranteeing an improvement in the quality of life (Ebele et al. 2017). This same concept was reported as pseudo-persistent compounds due to their consumption, properties, and resistance in aquatic environments (Im et al. 2020).

Anti-inflammatory drugs are called "pseudo-persistent" compounds, such as diclofenac (Khetan and Collins 2007; Magureanu et al. 2015), a contaminant commonly found in aquatic environments, with its degradation elucidated in several studies (Mugunthan et al. 2019; Rueda-Salaya et al. 2020).

Another non-steroidal compound widely found in water bodies is ibuprofen (Wang et al. 2019). This anti-inflammatory, analgesic, and antipyretic medication is consumed for medical and veterinary health (Wang et al. 2019). Khedr et al. found that different proportions between the anatase/brookite TiO_2 (A/B TiO_2) phases synthesized by the hydrothermal process are capable of degrading 100% of ibuprofen over 5 h (Khedr et al. 2019).

Akkari et al. synthesized photocatalysts based on the assembly of ZnO nanoparticles on the surface of sepiolite (Sep-clay material) and evaluated the degradation of pollutants such as ibuprofen, acetaminophen, and antipyrine under sunlight. The ZnO/Sep heterostructure exhibited improved photocatalytic performance compared to ZnO/SiO-Sep materials, with ibuprofen being the target compound with the highest degradation rate (Akkari et al. 2018).

Carbamazepine (CBZ) is an antiepileptic drug highly persistent and resistant to biodegradation, whose global consumption is around one thousand tons per year. Harboune et al. and Martinez et al. studied the CBZ photodegradation catalyzed by TiO_2 and ZnO, identified intermediates, and elucidated its degradation mechanism under UV-C/Vis irradiation (Martínez et al. 2011; Haroune et al. 2014). Caregnato et al. found that Ce-doped ZnO efficiently removed 53% of CBZ within 180 min under visible light irradiation (λexc. ≥ 475 nm) (Caregnato et al. 2021).

A huge variety of pharmaceutical products, including synthetic, anti-inflammatory, and antidepressant hormones have been detected by advanced analytical techniques in soils, surface waters, sediments, groundwater, and marine ecosystems (Gaw et al. 2014). Thus, numerous classes of pharmaceutical compounds are normally found contaminating waters (Magureanu et al. 2015).

The actions of antibiotics are essential due to the resistance of bacteria and impacts on fish fertility, which further reinforces the concern about the endocrine potential of these

Table 1 Photocatalysts based on ZnO applied in the removal of dyes, drugs and pesticides

Photocatalysts	Contaminants Wavelength and/or light source	Some details of the degradation	ZnO (He et al. 2019)
ZnO	Cephalexin/Xenon lamp	96%	(He et al. 2019)
Ce-ZnO	Nizatidine, Levofloxacin and Acetaminophen Mercury lamps	~95%	(Al Abri et al. 2019)
Ce-ZnO	Carbamazepine/UV/Vis lamps	94% (UV lamps)/53% (Vis lamps)	(Caregnato et al. 2021)
CuO/ZnO	Amoxicillin/Solar light	90%	(Belaissa et al. 2016)
Nd_2O_3/ZnO-GO	Ciprofloxacin/Mercury lamps	98.7%	(Arunpandian et al. 2019)
Fe Doped ZnO	Ciprofloxacin/Solar irradiation	66%	(Das et al. 2018)
ZnO/SiO_2/Fe_3O_4	Diazinon/*not specified*	95%	(Rezaei et al. 2019)
Cu–ZnO	Monocrotophos/*not specified*	~90%	(Hanh et al. 2019)
ZnO/SnO_2	Triclopyr/UV-lamp	~97%	(Yadav et al. 2019)
ZnO–Bi_2O_3	Lambda-cyhalothrin/Halogen lamp	89.91%	(Premalatha and Rose Miranda 2019)
ZnO NPs	Rhodamine B/Solar light—UV light	94.24% of RhB in 100 min in UV light and 93% in 180 min in solar light	ZnO NPs(Luque et al. 2022)
SiO_2/ZnO	Methylene blue	10 wt.% ZnO—90% / 40 min	(Rangelova et al. 2022)
ZnO/Vanadium/Neodymium	Rhodamine B—Methyl Orange / UV radiation	4% Nd-V-ZnO 99.3%—150 min	(Alam et al. 2019)
Au-ZnO core–shell	Methylene blue Methylorange / Xenon lamp—UV	25 Mm—MB 0.2 to 3 g/L (82% to 96%) for 80 min	(Verma et al. 2019)
ZnO/Graphene Oxide	Congo Red and—Eosin Yellow / Visible light	ZnO (EY—120 min = 31%) ZnO/RGO (EY—pH-4 = 57%) ZnO/RGO (EY—pH-19 = 98%)	(Ravi et al. 2018)
ZnO	Cephalexin / Under simulated sunlight	ZnO nanowires can efficiently photodegrade cephalexin even after 5 use cycles	(He et al. 2019)
ZnO nanorods (NRs)	Dyes and different pharmaceutics Under simulated sunlight	ZnO-S2 nanorod showed efficient photocatalytic degradation of different pharmaceuticals	(Mohammed et al. 2020)
ZnO	Temephos/Under sunlight	Photocatalytic ZnO films completely degraded Temephos pesticide under sunlight	(Serrano-lázaro et al. 2020)
Cu–ZnO (Hanh et al. 2019)	Monocrotophos/Visible light	Cu–ZnO exhibited excellent degradation of monocrotophos pesticide. Optimal pH = 7	(Hanh et al. 2019)
ZnO/SnO_2	Triclopyr Philips TL-D Actinic BL 15 W as light source	ZnO/SnO $_2$ > pure ZnO or SnO_2 10% of SnO_2 (ZS-2) exhibited highest photo catalytic activity towards degradation of TC	(Yadav et al. 2019)

drugs (Overturf et al. 2015; Rosendo et al. 2020). Due to health and environmental risks ranging from antimicrobial resistance, endocrine disruption, toxicity, and carcinogenicity, triclosan (TCS) is classified as an emerging contaminant with antimicrobial and fungicidal effects, widely used in the composition of personal hygiene products (Kosera et al. 2017; Azarpira et al. 2019). Kosera et al. (2017) evaluated the degradation capacity of triclosan using zinc oxide (ZnO) immobilized in the sodium alginate biopolymer. They found that the preparation of immobilized ZnO in biopolymer is an interesting alternative with cost–benefit, considering that biodegradable substrates are attractive from the environmental point of view and improve degradation (90%—under solar irradiation)(Kosera et al. 2017).

Pesticides play an important role in agriculture, helping control and prevent plant diseases and pests (Rousis et al. 2017; Jørgensen et al. 2019). Pesticides are considered emerging contaminants due to their high toxicity, low biodegradability, and environmental persistence (Evgenidou et al. 2005; Bachmann Pinto et al. 2018). The main groups of agricultural pesticides can be categorized by mode of action and chemical composition and their target organism (Huang et al. 2018; Mojiri et al. 2020). According to Ali et al. (2019), less than 1% (representative) of the applied pesticides reach their target organisms, and most of them remain in the soil and water. (Ali et al. 2019).

Herbicides represent about 46% of all global markets (Balmer et al. 2019) and are used in agriculture to control weeds (Harrington and Ghanizadeh 2017). Fungicides are often used in agriculture as prophylaxis to prevent diseases (Santísima-Trinidad et al. 2018). These bioactive and toxic substances directly or indirectly influence the soil's productivity and the agroecosystem's quality, possessing several modes of action and can have a wide range or be limited to a specific group of fungi (Mojiri et al. 2020). Insecticides are widely used to control insect pests, and their environmental safety remains a constant concern (Mojiri et al. 2020).

A chlorinated and highly persistent pesticide in the environment is lindane. The degradation was evaluated using Zn@ZnO CS nanocomposites as a photocatalyst (Jung et al. 2018). The authors pointed out that the hydroxyl radical ($^{\bullet}$OH) was the main reactive species for the degradation of lindane, which reached about 99.5% in 40 min under UV light. In addition, they conclude that defects, such as oxygen vacancies, can act as electron acceptors avoiding the process of recombination between charge carriers (Jung et al. 2018).

In this sense, due to the growing concern about the presence of these contaminants in the environment and their possible effects, researchers are increasingly investigating the degradation of pollutants using Zn-based photocatalysts Table 1.

Final remarks

Concomitantly, associated with concern about environmental impacts and the need for a better understanding of the occurrence and risks in water quality. This work evaluated the photocatalytic potential in the degradation of organic contaminants as an alternative to intense research in treating pollutants. The study showed that ZnO has excellent potential in the recovery of wastewater after breaking complex structures of contaminants into by-products of lesser aquatic impact, combined with photocatalysis and its various operational factors, ensuring safety for the health and well-being of people.

The reported experiments showed that the main limitation of ZnO is still to absorb only in the UV region, requiring structural modifications through effective methodologies to improve the quality of photocatalysts and their use in the visible region. Another challenge still observed is the recovery of photocatalysts for reuse, since most of the photocatalysts of ZnO and derivatives were in the form of powders. Among the main factors that affect the performance of ZnO and derivatives are parameters such as pH, the concentration of photocatalyst, concentration of the pollutant, source, and intensity of light, and others. Dyes are considered the most studied villains and are reported with effective/reliable results. However, few studies address degradation based on identifying photogenerated by-products and the proposal of appropriate mechanisms paths. There are several difficulties in developing stable photocatalysts and promising technologies due to the complexity of action plans and critical strategies for minimizing the cost–benefit of these materials (Ani et al. 2018).

Therefore, this review enhances points of scientific impact approaches, in particular the significant production and advances in the state of the art of ZnO, its structural and physical–chemical properties that represent principles of Green Chemistry, due to its non-toxicity, facile synthesis and chemical modification, low-cost and, standing out in the direct contribution to new compounds and the improvement of processes in environmental remediation.

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