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REVIEW

A Review on Nanomaterial as Photocatalysts for Degradation of Organic Pollutants

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Abstract

Eliminating hazardous organic contaminants from water is a major concern today. Nanomaterials with their textural features, large surface area, electrical conductivity, and magnetic properties make them efficient for the removal and photocatalytic degradation of organic pollutants. The reaction mechanisms of the photocatalytic oxidation of common organic pollutants were critically examined. A detailed review of articles published on photocatalytic degradation of hydrocarbons, pesticides, and dyes was presented therein. This review seeks to bridge information gaps on the reported nanomaterial as photocatalysts for the degradation of organic pollutants under sub-headings, nanomaterials, organic pollutants, degradation of organic pollutants, and mechanisms of photocatalytic activities.

Keywords Photocatalytic degradation · Organic pollutants · Nanomaterials

Introduction

Water is an essential resource for life on earth; it is a known fact that fresh water is an important necessity for our health. Recent population growth and globalization have resulted in a rapid spread of industries and, as a result, environmental pollution (Bayode et al. 2020, 2021). In recent times, much attention has been shifted to the release of toxic organic pollutants such as synthetic dyes, pharmaceuticals, steroid estrogens, pesticides, etc. into the water bodies. Synthetic dyes are among well-known occurring organic pollutants, it has been reported that about 280,000 tons of dyes are discharged in industrial effluents annually (Jin et al. 2007; Kubiak et al. 2019; Ramzan et al. 2019). A report by Adeel and his colleagues (2018) revealed that the world's human population of about 7 billion discharges approximately 30,000 kg/yr of natural steroid estrogen and an additional 700 kg/yr of synthetic estrogens solely from the practice of birth control using pills. The introduction of these pollutants into the water bodies can be direct through the disposal of untreated industrial wastes or indirectly through anthropogenic activities, farm run-offs, and wastewater treatment plants.

These contaminants are detrimental to aquatic ecosystem quality and human health, impairing recreational activities and other uses of water (Wang and Yang 2016). Some of

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these organic pollutants are classified as persistent organic pollutants (POPs) because they bio-accumulate and are not degradable when they enter water bodies. These organic pollutants can easily travel from the point of contamination to a pristine area or where they have never been produced (Krithiga et al. 2022).

The treatment of wastewater has recently been the subject of numerous scientific investigations. The removal of multi-component contaminants from water resources requires the utilization of efficient water treatment techniques. The wide usage of biological and physicochemical treatment processes in the industry is due to the simplicity of design as well as the enhanced remediation capacity associated with the techniques (Crini and Lichtfouse 2019). Nevertheless, the higher energy consumption associated with these conventional water treatment technologies has been a major impediment in the industry (Maktabifard et al. 2018). Finding a single treatment method that completely covers the efficient removal of all varieties of organic pollutants has become challenging due to the complexity and variety of organic chemicals used. This led to the development of alternative treatment technologies that will ensure the removal of recalcitrant organic pollutants. Some of these advanced treatment techniques comprise UV-illuminated processes, membrane-based processes, and advanced oxidation processes (Abdel-Fatah 2018; Boulkhessaim et al. 2022; Gaur et al. 2022). The use of non-destructive physical techniques including flocculation, reverse osmosis, and adsorption on activated charcoal results in secondary pollution because the contaminants are only transferred to new media, hence, there is a need for an effective environmentally benign method for the removal of these pollutants. The main advantage offered by advanced oxidation processes is that it ensures the effective degradation of organic pollutants via the generation of highly reactive hydroxyl radical, as opposed to conventional treatment processes that only ensures the physical transformation of pollutants without necessarily degrading them (Cheng et al. 2016; Krishnan et al. 2017). Among advanced oxidation processes, photocatalytic degradation has proven to be a quick, cost-effective, and environmentally benign method of removing persistent contaminants from water (Khan and Pathak 2020; Samuel et al. 2023).

Several recent studies have described the photocatalytic degradation of organic pollutants using different photocatalysts (Mei et al. 2022; Rostami et al. 2022; Silvestri et al. 2022; Subhiksha et al. 2022; Tang et al. 2022; Wang et al. 2022; Kanakaraju and Chandrasekaran 2023; Pattnaik et al. 2023). However, these studies have been singularly focused on one particular kind of photocatalyst such as metal-based photocatalysts or natural minerals. The present review was designed to focus on the use of nanomaterials as photocatalysts in the degradation of organic pollutants. Here, we described metal-based nanomaterials, carbon-based nanomaterials, and semi-conductor nanomaterials. In addition, the application of these nanomaterials in the removal of hydrocarbons, pesticides, and dyes was elucidated. Finally, the mechanisms by which photocatalysis occurs were illustrated.

Nanomaterials

Nanomaterials are defined as materials with a size or one of their dimensions that falls within the range of 1 to 100 nm (Laurent et al. 2010; Naseem and Durrani 2021). The unique properties of nanomaterials when compared to their bulk counterparts are significantly different, and their size-dependent effects become more noticeable at the nanoscale level. Surprisingly, by changing the shape and size at the nanoscale level, nanomaterials produce a distinct character with new features and powers (Kolahalam et al. 2019). Nanomaterials occur in different shapes like nanorods, nanosheets, spherical, oval, cubic, cluster, flower, triangular, needle-like, branched, etc. as illustrated in Fig. 1. Based on their form, size, characteristics, and constituents, nanomaterials also exist in different forms. These include polymeric nanomaterials, lipid-based nanomaterials, semi-conductor nanomaterials, metal nanoparticles, and carbon-based nanomaterials.

Metal-based Nanomaterials

Metals of trivalent and divalent ions are used as the building blocks for the creation of metal oxide nanoparticles (MONPs). They can be prepared using different techniques such as chemical and photochemical, due to their excellent electrical, optical, magnetic, and catalytic properties MONPs are useful in many fields like photocatalysis (Khalafi et al. 2019), catalysis (Jalpa et al. 2019), sensors, and heavy metal removal (George et al. 2018). Metal oxide nanoparticles (NPs) have attracted a lot of attention in many fields of science like chemistry, physics, and material sciences (Panji et al. 2016). For instance, Zinc oxide has good photocatalytic activity because of its unique properties like large surface area, super oxidative capability, high electrochemical stability, and low toxicity enable it to have a good capacity for the adsorption of small molecules. According to Naseem and Durrani (2021), TiO_2 is the most exceptional photocatalyst, it has low selectivity, making it ideal for degrading a variety of pollutants like polycyclic aromatic hydrocarbons (Guo et al. 2015), chlorinated organic compounds (Ohsaka et al. 2008), dyes (Lee et al. 2008), phenols (Nguyen et al. 2016), Macro and micron size plastics (Nabi et al. 2021), Cyanide (Chiang et al. 2002), Nitrophenols (Augugliaro et al. 1991), Chlorpyrifos, Cypermethrin, and Chlorothalonil (Affam and Chaudhuri 2013).

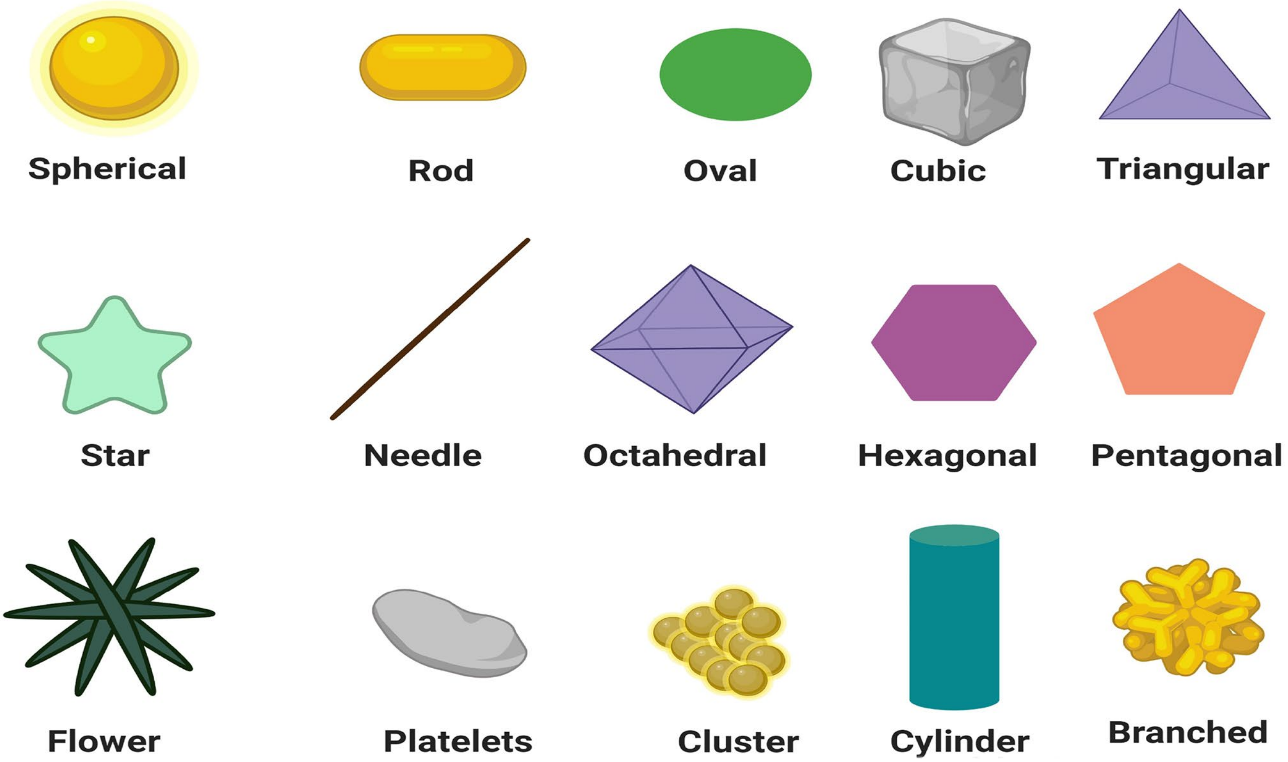

Fig. 1 Different shapes of nanoparticles. (Adapted from Hamida et al. 2020)

Carbon-based Nanomaterials

Carbon-based nanomaterials have recently attracted significant attention due to their outstanding physicochemical features like large surface area, excellent acid stability, and thermal resistance that make them viable candidates for a variety of many applications (Liu et al., 2022; Son et al., 2021). Carbon-based nanomaterials are classified according to their geometrical structure. Carbon nanostructures include particles that might be tube-shaped, horn-shaped, spherical, or ellipsoidal. Nanoparticles having the shape of tubes are called carbon nanotubes; horn-shaped particles are nanohorns, and spheres or ellipsoids belong to the fullerene group (Zaytseva & Neumann, 2016). Some of the shapes of carbon-based nanomaterials are shown in Fig. 2.

Carbon nanotubes (CNTs) are of two types i.e. single walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). They have high strength, favorable electron affinities, and good electrical conductivity (Scida et al., 2011, Zhao et al., 2015). The carbon allotropes known as carbon nanotubes (CNTs) are made of graphite and are in tabular form. The outside diameter of the tubes ranges from 3 to 30 nm and had at least two layers. Electrically, Single-walled carbon nanotubes can be further divided into metallic and semiconducting SWCNTs (s-SWCNTs and m-SWCNTs), whereas multi-walled carbon nanotubes mostly exhibit metallic behaviour. CNTs have unique and beneficial characteristics that make them suitable for a variety of electronic, biomedical, and other industrial applications. These characteristics include distinctive optical characteristics, high thermal, low cost, high surface area, high mechanical strength, and electrical conductivity (Chavali and Nikolova, 2019).

Semiconductor Nanomaterials

A wide range of various substances are used to create semiconductor nanocrystals (NCs). They are known as II-VI, III-V, or IV-VI semiconductor nanocrystals, depending on the periodic table groups into which these elements are formed. They exhibit both metallic and non-metallic qualities. They also have wide band gaps, modifying them demonstrates varied properties and exhibits large band gaps. They are commonly used in photocatalysis, electronic devices, drug delivery, solar cells, and environmental remediation (Fang et al. 2020; Sahu 2019).

Applications of Nanomaterials

Unique properties of nanoparticles like mechanical strength, large surface area, optically active, electronically active, high thermal conductivity, and chemically reactive make them

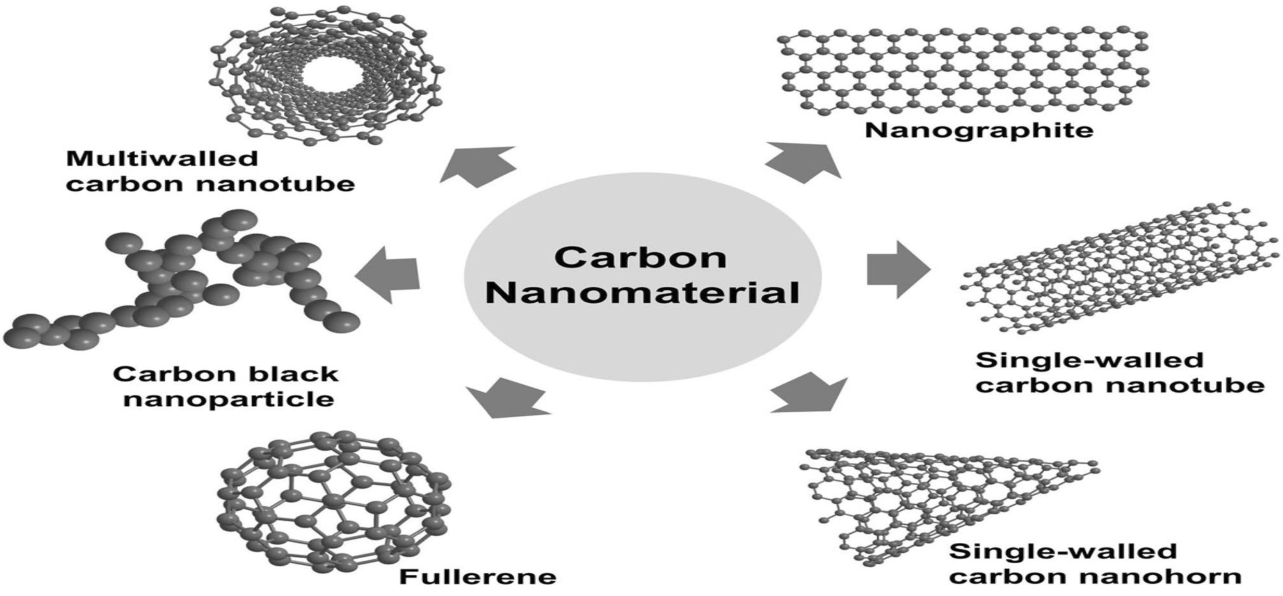

Fig. 2 Different shapes of carbon based nanomaterials. (Adapted from Yuan et al. 2019)

suitable for various applications. Nanomaterials have been used in the development of nanodevices that are found useful in pharmaceutical and biomedical applications (Loureiro et al. 2016; Martis et al. 2012; Nikalje 2015). Iron oxide particles such as magnetite (Fe_3O_4) or its oxidized form haematite (Fe_2O_3) are the most useful for biomedical applications (Ali et al. 2016). They are also used in wastewater treatment, due to their high surface-to-mass ratio, natural nanoparticles (NPs) are crucial in the solid/water partitioning of pollutants as they can either be absorbed onto their surface, co-precipitated during the synthesis of natural NPs, or trapped by NPs that had contaminants adsorb to their surface. (Khan et al. 2019). They are used as sensors, biosensors, and in the modification of electrodes to enhance their performance capability. NPs are found useful in energy storage devices, and also applicable in the form of electrodes (Li et al. 2019), however the importance of nanomaterials in the removal of organic contaminants cannot be overemphasized (Veisi et al. 2019).

Organic Pollutants

Organic pollutants are toxic synthetic chemical compounds that can persist in the environment over a long period of time. When their levels exceed the permissible limits, they become poisonous molecular compounds that can affect humans and cause many diseases. The main sources of these organic compounds are industrial items such as petroleum hydrocarbons, detergents, plastics, organic solvents, dyes, and insecticides. Some of these pollutants can resist degradation and bio-accumulate and remains for decades, they are classified as persistent organic pollutants (POPs). The salient features of POPs include the followings.

1. Persistence; POPs are not easily degradable either by chemical, physical, or biological degradation. They can retain in the soil, water, and air for decades.
2. Bioaccumulation; they build up inside the body to a point where they could be dangerous to both the environment and human health.
3. The ability to travel vast distances; POPs can travel through environmental media to far locations where they have never been utilized or produced, including the Arctic regions.
4. POPs are very hazardous and endanger both human and environmental health (Alharbi et al. 2018). It has been reported that some health-related issues like reproductive defects, preterm and immune toxicity are associated with exposure to organochlorine pesticides (Dalvie et al. 2004; Longnecker et al. 2001; Oyekunle et al. 2021), diseases like cancer, obesity e.t.c is found to associate with Polychlorinated biphenyl (Penell et al. 2014; Roos et al. 2013), also many PAHs have been reported to possess carcinogenic and genotoxic properties (Oyekunle et al. 2019; Rengarajan et al. 2015).

Hydrocarbons

Hydrocarbon contamination in the environment is a very serious issue whether it comes from petroleum, insecticides, or other hazardous organic materials. Being poisonous to all forms of life, petroleum hydrocarbons raise serious concerns about environmental pollution. Polycyclic aromatic hydrocarbons are organic pollutants that are typically colorless, white, or light-yellow solid substances composed of two or more fusions of carbon and hydrogen aromatic rings (Abdel-Shafy and Mansour 2016; Suman et al. 2016). The highest water solubility is seen in low molecular weight PAHs compounds like naphthalene, acenaphthene, and acenaphthylene while solubility declines with increasing molecular mass. They are categorized as semi-volatile chemicals because they have a low vapour pressure. However, as molecular weight increases, so do their boiling and melting points. The International Agency for Research on Cancer (IARC) has identified sixteen (16) PAHs that pose serious risks to human health because of their propensity to cause cancer and mutagenesis. These are Acenaphthene, Benzo(k)fluoranthene, Naphthalene, Benzo(a)anthracene, Chrysene, Benzo(a)pyrene, Acenaphthylene, Fluoranthene, Dibenzo(a,h)anthracene, Benzo(b)fluoranthene, Benzo(ghi)perylene, Phenanthrene, Benzo(j)fluoranthene, Indeno(1,2,3,d)pyrene, Anthracene, and Pyrene (European Union 2005; Keith and Telliard 1979; Yun et al. 2017; Zhang et al. 2020).

Pesticides

A pesticide is any substance that is used to eradicate, deter, or restrain specific plant or animal life forms that are regarded as pests. Most of the chemicals used in the production of pesticides belong to the family of carbamates, organochlorides, pyrethroids, organophosphates, and other substances (Intisar et al. 2022). Among these classes of pesticides, organochlorine is the most dangerous with deleterious effects on human health (Rani et al. 2017). An organochlorine compound is an organic compound that has at least one chlorine atom covalently attached to it as the main functional component. They have a wide range of applications due to their wide structural variety and divergent chemical properties, OCPs are chlorinated hydrocarbons that were extensively used in agriculture and mosquito control from the 1940s to the 1960s. Examples of compounds in this group include DDT, DDE, methoxychlor, Lindane, Endosulfan, Chlordane, Diclof-methyl, Dieldrin, toxaphene, mirex, Aldrin, DDD, and Benzene hexachloride. Up until 1980, the use of these organochlorine pesticides (OCPs) was extremely successful; however, suggestions for controlled usage of OCPs were made due to the serious health risks they posed, including the potential for cancer, endocrine disruption, immune system disorders, reproductive issues, and other chronic diseases as a result of their persistence or strong resistance in ambient environmental conditions. (Augustijn-Beckers et al. 1994; Kumar et al. 2013; Syafrudin et al. 2021).

Dyes

Dyes are compounds that give color to a surface when applied through a process that modifies if only temporarily, the crystal structure of the colored substances (Bafana et al. 2011). They are substances, both natural and artificial that add colour to the world and enhance its beauty.

A group of organic chemicals known as textile dyes are frequently regarded as contaminants and are mostly released into wastewater as a result of chemical textile finishing processes. According to estimates, over 10,000 different types of dyes and pigments are utilized in industry, and more than 7 million tons of synthetic dyes are produced annually around the world (Ogugbue and Sawidis 2011; Robinson et al. 2001). Azo dyes are the most used dyes and account for more than 60% of total dyes (Gürses et al. 2016; Shah 2014) and approximately 70% of all the dyes used in the industry are azo dyes (Lipskikh et al. 2018; Berradi et al. 2019). Azo compounds have been used in a variety of scientific and industrial fields, including color waxes, oils, gasoline, solvents, polishes, paper, varnish, food, leather, plastics, cosmetic medicine, and automobiles (Al-Khuzaie and Al-Majidi 2020; Dixit et al. 2007; Guerra et al. 2018; Patel and Dixit 2014).

Additionally, they are employed as antibacterial, anti-diabetic, and anti-tumor agents (Adu et al. 2020; Rabbani et al. 2020).

Photocatalytic Degradation of Organic Pollutants Using Metal Oxide Nanomaterials

Photocatalysis is the process that takes place when a light source interacts with the semiconductor components that make up the photocatalyst on its surface (Adeola et al. 2023; Alegbeleye et al. 2022; Ore et al. 2023). However, this process depends on in-situ photogenerated hydroxyl radicals (OH^-), superoxide radicals (O_2), and positively charged holes (h^+) which completely degrade organic contaminants. Oxidation from photogenerated holes and reduction of photogenerated electrons are the two simultaneous processes that take place during this process (Adeola et al. 2022). Consequently, the process of removing impurities by photocatalysis is effective, ecologically benign, inexpensive, and simple (Zhang et al. 2018). Jiang et al. (2015) synthesized an aggregate silver oxide nanoparticle that demonstrated excellent photocatalytic performance in both artificial and natural light. According to the result, methyl orange decomposed under near-infrared light in 40 min and under sunlight, artificial ultraviolet, and visible light in 120 s. Fe_3O_4 mesoporous carbon shell was used by Angamuthu et al. (2017) to develop nanomaterial that was used to degrade the dye methylene blue, with this artificial nanomaterial, methylene blue dye was degraded with excellent catalytic activity. In 2019, Kubiak and his colleagues published a paper on the synthesis of extremely crystalline photocatalysts based on TiO_2 and ZnO for the oxidation of organic contaminants. The created TiO_2-ZnO binary oxide systems exhibit remarkable photodegradation efficiency of organic contaminants of 90%. In their 2018 study, Harun et al. (2018) examined the effectiveness of photocatalytic degradation in the absence and presence of a photocatalyst as well as the impact of a light source for the decolorization of Congo red dye under solar and UV light, TiO_2 was the catalyst utilized. Sunlight and artificial UV light both have a 30 min degradation rate of the dye of up to 64.72% and 66.99%, respectively. Photocatalytic oxidation of phenol was studied by Hayat et al. (2011), it was reported that the Photocatalytic degradation efficiency of phenol was 97% using nano NiO and UV laser irradiation was accomplished in a short amount of time as compared with conventional setups like lamps. Jassal and his coworkers (2015) reported photocatalytic degradation of Eriochrome Black T (EBT) and Malachite MG) Green with degradation efficiency of 94.15% and 76.13% for MG and EBT respectively using nanocubes. Ullah and Dutta (2008) used Mn doped ZnO NPs for photodegradation, and their photocatalytic effectiveness was determined by the degradation of aniline and MB dyes under visible light from a tungsten lamp. It was reported that ZnO-Mn^{2+} NPs can be employed as a better photocatalyst than undoped ZnO since Mn doped ZnO demonstrated a 50% higher degradation rate than undoped ZnO. The potential of cobalt and cobalt oxide nanoparticles as nanocatalysts for the degradation of murexide and EBT dye in wastewater in the presence of sunshine was investigated by Adekunle et al. (2020). The highest degradation efficiency was reported for chemically synthesized Co-nanoparticles (43.6%) toward murexide dye at 25 mg loading and 40 min exposure time, while the highest degradation efficiency for microwave-synthesized Co_3O_4 nanoparticles (39.4%) was reported for EBT at 10 mg loading and 40 min sunlight exposure time. Adekunle and his colleagues worked on a study comparing the photocatalytic degradation of dyes in wastewater utilizing solar-enhanced iron oxide (Fe_2O_3) nanocatalysts made by chemical and microwave processes. According to the research, microwave-produced Fe_2O_3 nanoparticles had the highest degradation rates for the dyes Murexide (98%) and EBT (96%) at a 25 mg loading and 40 min exposure, whereas chemically produced Fe_2O_3 nanoparticles had the highest degradation rates for the dye Murexide (15.2%) at a 5 mg loading and 30 min sunlight exposure. With 15 mg of catalyst loading and 40 min of exposure, the chemically produced Fe_2O_3 nanoparticles in EBT degraded at a rate of 21.4%. (Adekunle et al. 2021). The mechanism of the degradation of EBT and murexide is illustrated in Eqs. (1)–(4) below.

$$Fe_2O_2 + hV \rightarrow Fe_2O_3(e^- + h^+) \tag{1}$$

$$\mathrm{h}^+(\mathrm{VB}) + H_2\mathrm{O} \rightarrow \mathrm{OH}^{\cdot} \tag{2}$$

$$\mathrm{e}^-(\mathrm{CB}) + \mathrm{O}_2 \rightarrow \mathrm{O}_2^- \tag{3}$$

$$EBT/Murexide + h^+ + O_2^- + OH \rightarrow Degradation\ products \tag{4}$$

Photocatalytic Degradation of Organic Pollutants Using Carbon-based Nanostructures

Carbon-based nanostructures could be viewed as an ideal candidate for enhancing the photocatalytic effectiveness of nanoparticles by facilitating the transferring of photo-generated electrons and reducing charge recombination.

One report by Nguyen et al. (2018), utilized graphene oxide (GO) as a support for the deposition of ZnO NPs through a simple hydrothermal process, in which GO prevent ZnO particle from aggregation as well as the efficient separation of photo-generated electrons and holes (e^- and h^+) on the surface of the ZnO. The nanocomposite was successfully used for the degradation of methyl orange (MO) during which more than 95% of MO was decomposed at optimal conditions.

In a work reported by Atchudan et al. (2017) that involves the degradation of MO and MB using TiO_2-GO nanocomposite, a maximal degradation efficiency of 84% and 100% were obtained for MO and MB respectively by using a two-step sol-gel deposition technique. Durmus et al. (2019) synthesized a GO/ZnO nanocomposite catalyst and was successfully used as a photocatalyst to degrade the basic fuchsin (BF) dye. The degradation of BF dye, a model compound in an aqueous media, was found to be more effectively photocatalyzed by the nanocomposite structure because it decreased the band gap of ZnO nanoparticles. Khataee and his co-workers in their work on photocatalytic degradation of organic dyes illustrate how photogenerated electron-hole pairs by the synthesized nanoparticle facilitate the degradation of organic pollutants. It was observed that when TiO_2 is irradiated with light, an electron excites out of its energy level, leaving a hole in the valence band and electrons are promoted from the valence band to the TiO_2 conduction band, resulting in electron-hole pairs (Khataee and Kasiri 2010) as shown in Fig. 3 below.

Jo et al. (2017) worked on the photocatalytic degradation of oxytetracycline and Congo red using Cobalt-titanium

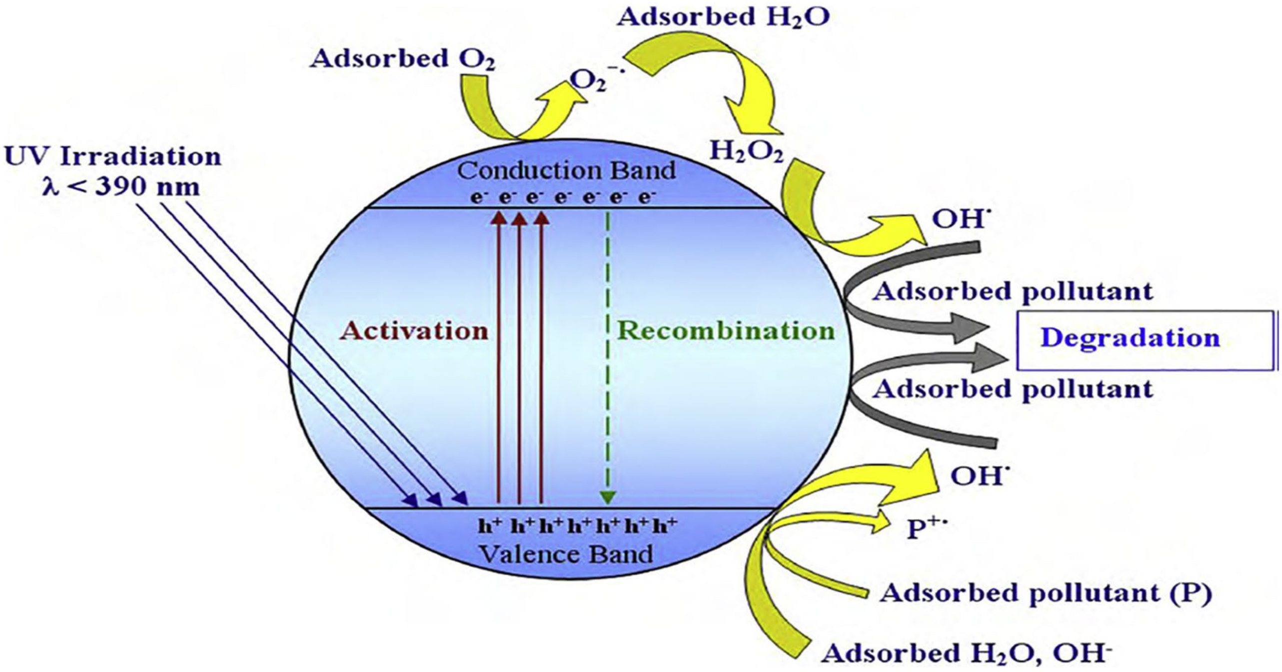

Fig. 3 General mechanism of the photocatalysis on TiO_2 nanomaterial. [Reprinted from Khataee and Kasiri 2010]

mixed metal oxides. It was discovered that the photocatalytic oxidative destruction of oxytetracycline and Congo red was greatly improved by heterojunction formation between a low concentration of discrete Co_3O_4 nanoparticles and anatase titania, which was further enhanced by the addition of trace Graphene oxide as shown in Fig. 4.

A list of different percentages of degradation of some organic pollutants by some nanomaterials as reported in some literature is given in Table 1 below.

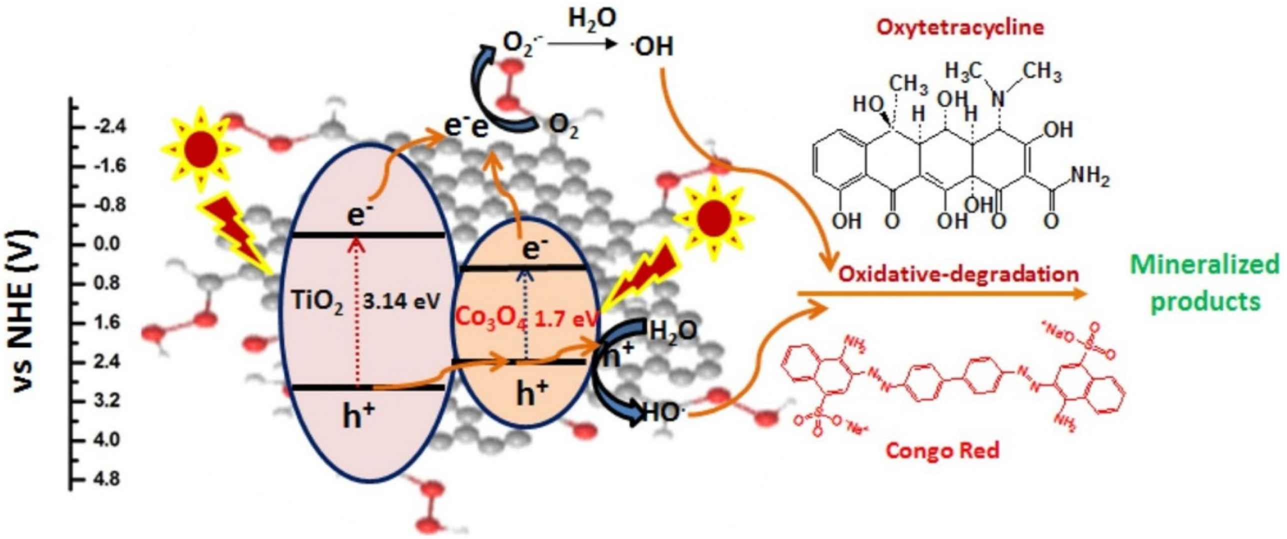

Fig. 4 Proposed mechanism for the photocatalytic degradation of organic pollutants by $Co_3O_4/TiO_2/GO$ nanocomposite. (Reprinted from Jo et al. 2017)

Table 1 Percentage degradation of some organic pollutants using nanomaterials

Organic pollutant	Catalyst	% degradation	References
EBT	Bi_2WO_6 NPs	74	Lai et al. (2019)
EBT	KZnHCF	76.13	Jassal et al. (2015)
MG	KZnHCF	94.15	Jassal et al. (2015)
EBT	CO_3O_4 NPs	39.4	Adekunle et al. (2020)
Paracetamol	TiO_2-G	88	Vaiano et al. (2018)
Malathion	Zn^{2+}-doped TiO_2	98	Nasseri et al. (2018)
EBT	SnO_2 NPs	94	Srivastava and Mukhopadhyay (2014)
EBT	$ZnFe_2O_4$ NPs	92	Ikramullah et al. (2020)
Metronidazole	$BiVO_4$ /N-rGO	95	Appavu et al. (2018)
EBT	ZnO	90	Kaur and Singhal (2015)
EBT	$BiFeO_3$ NPs	64.6	Khan et al. (2020)
Congo Red	$BiVO_4$ /N-rGO	95	Appavu et al. (2018)
EBT	Fe_2O_3	96	Adekunle et al. (2021)
Murexide	Fe_2O_3	98	Adekunle et al. (2021)
Chloramphenicol	$BiVO_4$ /N-rGO	93	Appavu et al. (2018)
Murexide	TiO_2 NPs - AC	98	Davoodi et al. (2014)
Methylene blue	$BiVO_4$ /N-rGO	98	Appavu et al. (2018)
Murexide	ZnO NPs	45.1	Alkhateeb et al. (2007)
Murexide	TiO_2 NPs	60.1	Alkhateeb et al. (2007)
Ciprofloxacin	$ZnO/ZnAl_2O_4$/rGO	90.6	Ni et al. (2018)
Murexide	$BiFeO_3$ NPs	83.4	Khan et al. (2020)
Phenol	Fe-Fe_3O_4-GO	87	Le et al. (2018)
Phenol	Fe_3O_4-GO	75	Le et al. (2018)
Endosulfan	TiO_2	99	Sivagami et al. (2016)
Chlorpyrifos	TiO_2	94	Sivagami et al. (2016)
Amido black dye	Co/TiO_2	90	Ali et al. (2018)
Azo dye	CdS	95	Senasu and Nanan (2017)
Methylene blue	NiO-ZnO	97	Rogozea et al. (2017)
Methylene blue	AKB	94	Zhou et al. (2018)
Rhodamine B	BiOI/N-doped rGO	83	Lu et al. (2018)
Remazol yellow RR dye	α-Fe_2O_3	77	Bhuiyan et al. (2020)
Acid blue 74 dye	Ag-Ag_2O-ZnO/GO	90	Umukoro et al. (2016)

Mechanism of Photocatalytic Activity

A chemical reaction known as photocatalysis is initiated by light and takes place when the compound comes into contact with photons with high enough energies to trigger free radical processes. The mechanism of organic pollutant photo-oxidation can take place in two ways, direct or indirect.

(a) Direct degradation mechanism

This type of photocatalytic degradation takes place under visible light because of the ease of absorption of some visible light. This mechanism involves the excitation from the ground state of the organic pollutant to the triplet excited state (organic pollutant) under visible light photons (> 400 nm). A further transformation of the excited state organic pollutant species into a partially oxidized radical cation (Dye^+) is accomplished by injecting an electron into the conduction band of the nanoparticle. The system-dissolved oxygen and these trapped electrons combine to produce superoxide radical anions (O_2^-), which in turn lead to the creation of hydroxyl radicals (OH^-). The oxidation of organic molecules is mostly caused by these OH^- radicals as represented by Eqs. (5) and (6) below.

$$\text{Dye} + \text{hv} \rightarrow \text{dye}^* \tag{5}$$

$$\text{Dye}^* + \text{semiconductor} \longrightarrow \text{dye}^+ + TO_2^- \tag{6}$$

The entire process can be summarized using the Fig. 5 below.

(b) Indirect dye degradation mechanism.

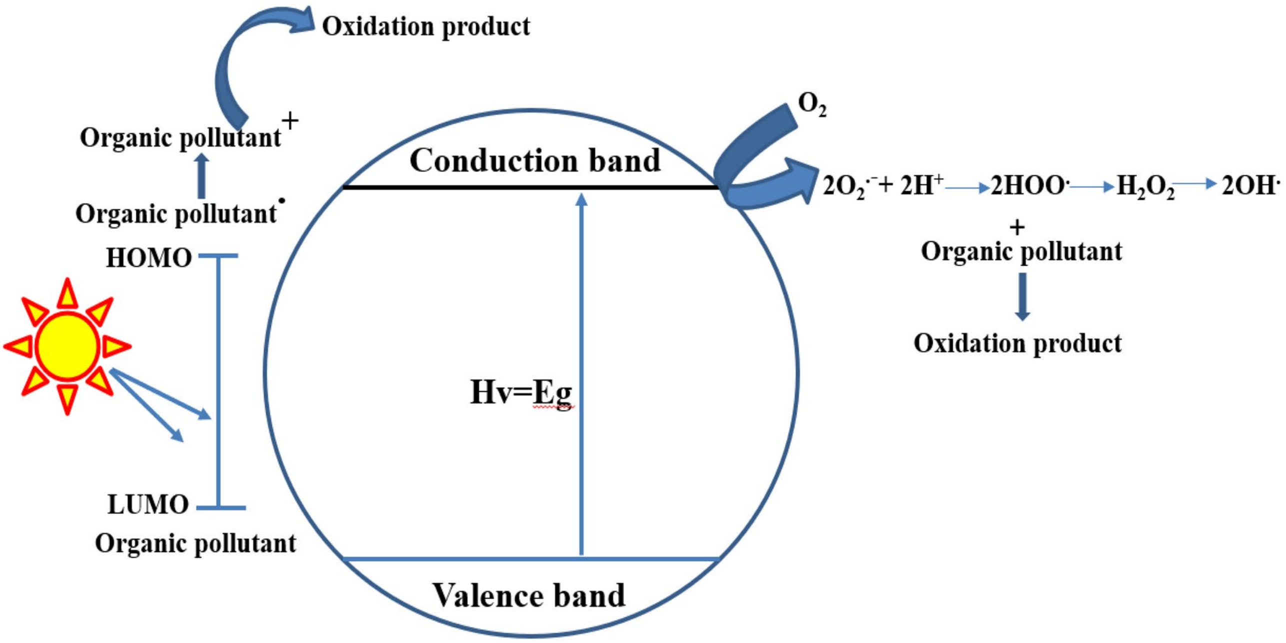

Fig. 5 Pictorial representation of direct organic pollutant degradation process. (Adapted from Ajmal et al. 2014)

The photocatalytic reaction starts when a photoelectron is promoted from the filled valence band of a semiconductor photocatalyst when it is exposed to radiation. The energy of the absorbed photon (hv) is equal to or higher than the semiconductor photocatalyst's band gap. A hole (h_{VB}^+) is left in the valence band because of excitation, consequently leading to the generation of an electron and hole pair (e^-/h^+) as illustrated in Eq. (7) below.

$$TiO_2 + hv(UV) \rightarrow TiO_2(e^-(CB) + h^+(VB)) \tag{7}$$

The water and the photogenerated holes in the valence band subsequently combine to form the hydroxyl radical (Eq. 8).

$$H_2O_{(ads)} + h^+(VB) \rightarrow OH^\cdot_{(adS)} + H^+_{(ads)} \tag{8}$$

Adsorbed organic molecules or those that are very near the catalyst surface are attacked non-selectively by the hydroxyl radical that forms on the irradiation semiconductor surface, and this causes them to mineralize to a degree depending on their structure and stability level.

While the photogenerated hole (h_{VB}^+) reacts with surface-bound water or OH^- to create the hydroxyl radical, the electron in the conduction (e_{CB}^-) is taken up by the oxygen to create an anionic superoxide radical (O_2^-), which may participate in further oxidation but also prevents electron-hole recombination, maintaining electron neutrality within the semiconductor molecule.

$$O_2 + e^-(CB) \rightarrow O^-_{2(ads)} \tag{9}$$

The hydroperoxyl radical (HO_2^-) that is formed from the superoxide (O_2^-) is protonated to form H_2O_2, which further dissociates into highly reactive hydroxyl radicals (OH^-).

$$O^-_{2(ads)} + H^+ \leftrightarrow HOO^\cdot_{(ads)} \tag{10}$$

$$2HOO^\cdot_{(ads)} \rightarrow H_2O_{2(ads)} + O_2 \tag{11}$$

$$\text{Organic pollutant} + OH^\cdot \rightarrow CO_2 + H_2O \quad \text{(organic pollutant intermediates)} \tag{12}$$

$$\text{Organic pollutant} + h^+(VB) \rightarrow \text{Oxidation products} \tag{13}$$

$$\text{Organicpollutant} + e^-(CB) \rightarrow \text{Reductionproducts} \tag{14}$$

Both oxidation and reduction processes occur frequently on the surface of the photoexcited semiconductor photocatalyst. The overall process of these reactions can be summarized using Fig. 6 below.

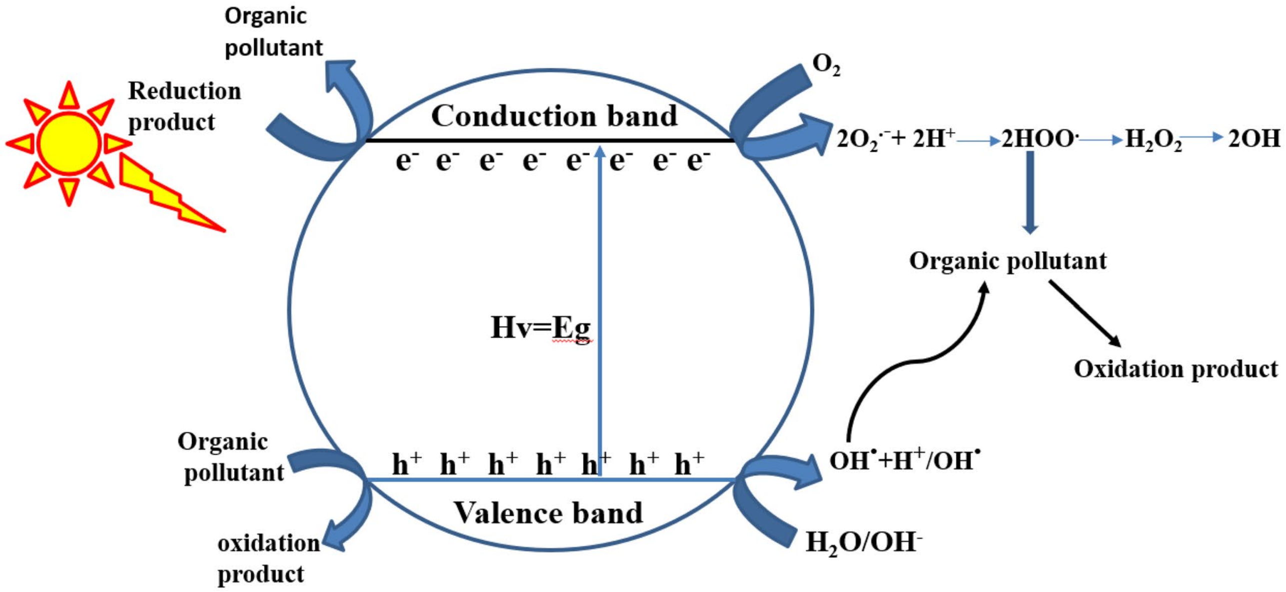

Fig. 6 Pictorial representation of indirect organic pollutant degradation process. (Adapted from Ajmal et al. 2014)

Conclusion

The increase in organic contamination of water bodies through anthropogenic activities is a major concern due to their bioaccumulation and adverse health effects on man and his environment. This paper reviewed studies addressing the photocatalytic degradation of organic pollutants. It was observed that strong oxidizing agents like hydroxyl and related radicals can break down any organic pollutant into smaller degradation products that are either practically innocuous or less toxic. Different organic contaminants like hydrocarbons, steroid estrogens, pharmaceuticals, dyes, pesticides, phenols, etc. can be photo-oxidized by hydroxyl radicals. Filtration, ion exchange, coagulation/flocculation, aerobic degradation, anaerobic degradation, ozonation, and photocatalytic processes are just a few of the techniques that have been utilized to remove organic pollutants from wastewater. As an alternative to physical, chemical, and biological processes, photocatalysis employing nanomaterials showed greater potential than the aforementioned techniques. Furthermore, nanocatalysts quickly oxidize and are cheap, chemically stable, and environmental-friendly. While these photocatalysts have shown relatively high degradation of the studied organic pollutants, it appeared that the efficiency of the process is largely driven by the choice of photocatalysts. The choice of photocatalyst equally depends on the target pollutant. There is significant variation in the ability of different photocatalysts to interact with specific pollutants and initiate essential chemical processes. Understanding the photocatalytic characteristics of various materials, as well as their interactions with certain pollutants, is therefore critical for achieving optimal degradation efficiencies. It is recommended that further research should be directed into the development of highly efficient photocatalytic materials for the abatement of these recalcitrant organic pollutants. Also, attention should be focused on improving the selectivity of nanomaterials for the removal of these pollutants so that photocatalytic technology can gain ground in industrial applications.

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