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# Dyes and their toxicity: removal from wastewater using carbon dots/metal oxides as hybrid materials: a review

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Clean water and a hygienic living environment are fundamental necessities for a healthy life. However, both domestic and industrial carcinogenic dyes have significantly reduced drinkable water sources and caused severe ecological problems. This situation seriously threatens human civilization, sustainable global development, and the marine ecosystem. Researchers have focused on advanced techniques to remove or degrade toxic dyes more effectively than conventional wastewater treatment methods to address these challenges. This study explores an efficient, quick, and cost-effective approach for degrading and removing toxic dyes from wastewater using carbon dots (CDs) and metal oxides (MOs). The hybrid CD/MO system offers several advantages over traditional methods, including enhanced optical properties, improved catalytic efficiency, structural modification, enhanced adsorption capacity, superior charge transfer, and increased chemical stability. This review provides a detailed introduction to dyes and their classifications, as well as the environmental and health impacts of dyes. It then describes various dye removal methods and, for the first time, explores the application of hybrid carbon dot/metal oxide systems for removing toxic dyes from wastewater. Additionally, we discuss current challenges and future perspectives in dye removal, aiming to provide an overview of recent advancements in wastewater treatment, highlight the associated challenges, and suggest potential future directions.

## 1. Introduction

Industrial wastewater is a major contributor to aquatic pollution, posing significant risks to ecological systems and human health.[1] Notably, printing and dyeing wastewater from the textile industry, characterized by high concentrations of chemical oxygen demand (COD), toxic substances, and colored pollutants,[2–5] is considered one of the most heavily polluted types of industrial effluent.[6] If released untreated, it can result in severe and unpredictable environmental consequences.[7,8] Thus, treating this wastewater to a safe level is critically important. Various treatment methods have been developed to address this issue, including electrochemical,[9] biodegradation,[10] coagulation,[11] chemical precipitation,[12] membrane filtration,[13] advanced oxidation processes,[14] and adsorption.[15] Among these, adsorption and degradation are often preferred due to their cost-effectiveness, ease of reuse, low waste, straightforward operation, and environmental sustainability.[16–20]

A wide range of materials has been developed for the effective removal of toxic dyes, including metal–organic frameworks (MOFs),[21] organic materials,[22] activated carbon,[23] covalent organic frameworks (COFs),[24] sulfur dots,[25] synthetic polymers,[26] and zeolites.[27] However, metallic materials often exhibit limited adsorption capacity due to their restricted number of accessible binding sites. Similarly, the dye adsorption efficiency of natural biopolymers like chitosan depends on pH levels. While materials such as MOFs, COFs, and synthetic polymers offer high adsorption capacity, their use is hindered by the high cost of raw materials and complex synthesis procedures.[28] Additionally, some materials, including activated carbon, zeolites, and organic materials, lack stability across a broad pH range, resulting in inconsistent and suboptimal performance under varying operating conditions.[29] Additional materials are still needed to meet the demand for dye removal.

Carbon dots (CDs), a new class of carbon-based nanomaterials, have become a major focus of research in recent years.[30–32] These zero-dimensional, quasi-spherical nanoparticles were discovered serendipitously during the purification of

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single-walled carbon nanotubes in 2004.[33] Since then, numerous CDs with varying sizes, structures, and photoluminescent properties have been synthesized. CDs are generally categorized into graphene quantum dots, carbon quantum dots, and polymer dots.[34]

Compared to other carbon materials, CDs stand out due to their excellent photoluminescent properties, diverse functional groups, abundance of raw materials, ease of modification, and straightforward synthesis methods.[35] Additionally, CDs are regarded as superior alternatives to traditional semiconductor quantum dots because of their high water solubility, low toxicity, excellent biocompatibility, and exceptional resistance to photobleaching.[35–41] These advantages make CDs highly attractive for a wide range of applications, including bioimaging,[42] drug delivery,[43] sensing,[44] catalysis,[45] and energy storage.[46]

Due to their shared characteristics, metal oxides, such as vanadium, chromium, titanium, zinc, tin, and cerium, exhibit similar primary photocatalytic processes. These processes include light absorption, which triggers charge separation and forms positive holes capable of oxidizing organic substrates.[47,48] When a MO is illuminated by UV light, visible light, or a mix of both, the absorbed energy excites electrons, causing them to transition from the valence band to the conduction band, creating an electron–hole pair ($e^-/h^+$). This photogenerated $e^-/h^+$ pair can reduce and/or oxidize compounds adsorbed on the photocatalyst's surface. The photocatalytic activity of MOs is primarily attributed to two mechanisms: (i) the generation of hydroxyl (OH) radicals through the oxidation of hydroxide ($OH^-$) anions, and (ii) the formation of superoxide ($O_2^-$) radicals by the reduction of molecular oxygen ($O_2$). These reactive species interact with pollutants, facilitating their degradation or transformation into less harmful byproducts.[49] Consequently, to broaden their potential applications, incorporating CDs into metal oxides is an effective strategy to modify and enhance their unique electronic, catalytic, and optical properties. Adding CDs significantly improves metal oxides' versatility and functionality.[50]

This review provides the first in-depth analysis of the fabrication of CDs hybridized with MOs and their applications in removing toxic dyes. It also covers the classification of dyes, their environmental and health impacts, methods of dye removal, and factors affecting dye degradation. Lastly, the review explores future opportunities and challenges in advancing CD/MO hybrid materials for efficient dye removal.

### 1.1. Motivation of this study

In the near future, fostering a sustainable ecosystem will ensure a healthier environment. Industrial discharges significantly contribute to environmental pollution, harming human health, aquatic organisms, terrestrial wildlife, and plant life. Among these pollutants, effluents from the dye industry stand out as major contributors to environmental degradation. This growing concern inspired and motivated us to undertake the task of authoring this review study. To promote environmental sustainability, this review highlights the potential of combining carbon dots and metal oxides as innovative approaches for removing dyes from wastewater. The article primarily focuses on defining various dyes, understanding the toxicity caused by water pollution, and addressing this issue using carbon dot- and metal oxide-based photocatalysts and adsorbents. By exploring these modern solutions, this study aims to contribute to developing cleaner technologies for mitigating dye pollution and protecting the environment.

### 1.2. Scope of this study

Various nanomaterials have been applied to environmental applications, including COFs,[51–53] MOFs,[54–57] CDs,[58] and sulfur dots.[59] Among them, CDs have shown particular promise when combined with metal oxides such as $TiO_2$, enhancing catalytic activity due to their unique properties, including an adjustable bandgap, efficient charge separation, and high carrier mobility. Integrating CDs with $TiO_2$ not only broadens the light absorption range but also creates additional active sites and enables better control over electron–hole recombination. This combination results in a material with excellent electrical conductivity, increased surface area, structural adaptability, superior thermal conductivity, chemical stability, and a reduced quantum-limited bandgap, all of which make it a strong candidate for environmental remediation.[60,61] This review focuses on recent advancements in the application of CD/MO hybrids for dye removal. It covers the toxicity and classification of dyes, their impacts on the environment and human health, the mechanisms of dye removal based on hybrid materials, and their wide range of applications using various metal oxides, such as titanium oxide, zinc oxide, iron oxides, and others. These insights contribute to a deeper understanding of the versatile role that CD/MO hybrids play as photocatalysts in wastewater treatment.

## 2. Dyes and classifications

Polluted water poses a significant threat to the environment, with widespread implications. Various pollutants, including insecticides, dyes, metal ions, chemicals, and inorganic substances, contribute to environmental contamination. The persistent and hazardous nature of dyes is primarily due to the stable aromatic rings in most dye structures. Even minimal amounts of dye can contaminate an ecosystem.[62] Consequently, dye pollution is among the most harmful contaminants that must be addressed. Most dyes resist degradation under light, heat, and oxidative conditions, making them non-biodegradable. Furthermore, dyes negatively impact aquatic ecosystems by diminishing the aesthetic quality of natural habitats.[63] Humans have been using dyes for thousands of years, with the earliest recorded use of colorants traced back approximately 180 000 years to Neanderthals. However, the first documented use of an organic colorant occurred much later, around 4000 years ago, when the blue dye indigo was discovered in the wrappings of mummies in Egyptian tombs. Until the late 19th century, most dyes were derived from natural sources such as plants, insects, and mollusks and were typically produced on a small scale. The large-scale production of synthetic dyes began after 1856, following Perkin's groundbreaking discovery of mauveine, the first synthetic dye.[64]

Dyes are vibrant compounds that impart color to materials like fabrics, paper, and other suitable surfaces. Their ability to adhere to compatible substrates enables this coloration process.[65,66] Humans have been using dyes for over a millennium in diverse applications. Historically, dyes were primarily produced on a small scale from naturally occurring sources such as plants and insects, referred to as natural dyes.[67] However, natural dyes had certain limitations, including a restricted range of colors and subdued shades that tended to fade with exposure to sunlight and repeated washing.[68] Dyes are a vital category of organic macromolecules that play a significant role in our daily lives and find extensive use across various industries. Industries such as textiles, paints, plastics, dye-sensitized solar cells, optics, metal extraction, and sensors rely heavily on dyes. Dyes are classified based on several characteristics, including their molecular structure, origin, color, and application methods, as outlined in the color index (CI).[69] Dye molecules consist of two main components: chromophores, which generate the color, and auxochromes, which not only support the chromophore but also enhance the molecule's water solubility and improve its ability to bind to fibers. Dyes have a wide range of structural variations and can be categorized in multiple ways.[64] Commercial dyes can be categorized in various ways, including structure, color, and application methods.[70] Due to the complexity of naming dyes based on their chemical structures, classification by application is often preferred. Synthetic dyes come in various types and are typically categorized based on their molecular structure; however, they can also be grouped by their application method or solubility. For instance, acid, direct, mordant, basic, and reactive dyes are soluble, whereas azo, disperse, sulfur, and vat dyes are classified as insoluble,[70] as depicted in Fig. 1.

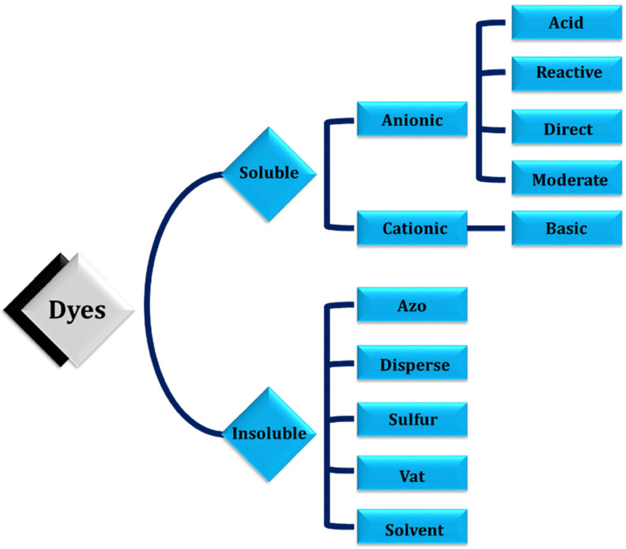

Fig. 1 Classification of dyes based on solubility.

## 3. Impacts of dyes on the environment and health

Colors are a part of our everyday lives, visible in the clothes we wear and the objects that surround us, whether from natural or artificial sources. Prior to the 19th century, natural resources were the sole source of colors. Plants, trees, and lichens were the primary contributors, with insects and mollusks playing a secondary role. Despite thousands of years of dye usage, only a few of these natural dye sources were used consistently, highlighting their instability. Today, the commercial market offers over 7000 colorants.[71] Samsami *et al.* (2020) noted that the textile industry is the largest contributor to dye release into the environment, accounting for 54% of discharges. Other significant sources include dyeing factories (21%), paper and pulp factories (10%), paint and tannery facilities (8%), and dye manufacturing plants (7%) (Fig. 2). Many dye mixtures are released during the textile production process.[72]

Basic dyes exhibit high color intensity and remain highly visible even at minimal concentrations.[73,74] Complex chromium-based dyes are often classified as carcinogenic.[75,76] Dyes can impair photosynthetic activity in aquatic organisms by reducing light penetration and may pose toxicity risks due to the presence of metals and aromatic compounds.[77,78] Additionally, dyes are known to be carcinogenic, mutagenic, and teratogenic, affecting various microorganisms and fish species. In humans, exposure to dyes can lead to severe health issues, including dysfunction of the kidneys, reproductive system, liver, brain, and central nervous system.[79,80] Azo dyes are particularly hazardous due to toxic amines in their effluents,[81] while anthraquinone-based dyes are highly resistant to degradation, retaining their color for extended periods in wastewater.[82] Reactive, water-soluble dyes result in 5–10% of their volume being discharged as highly colored effluent, causing significant environmental problems.[83] Their chemical stability and low biodegradability allow them to bypass conventional treatment processes, making their removal critically important.

Industrial dyes are widely used across various sectors but pose significant risks to human health and the environment. Below is a summary of the environmental impact of various toxic dyes.

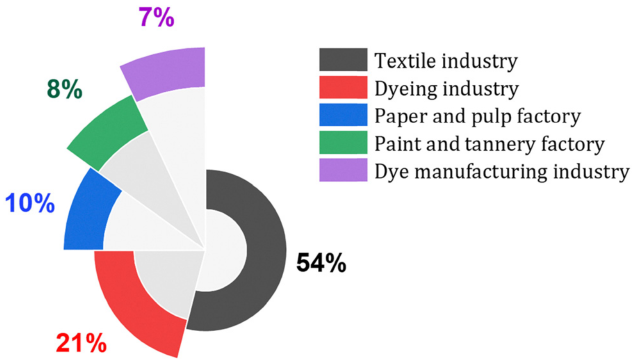

Fig. 2 Key industries responsible for releasing dyes into the environment.

Methylene blue, used in dyeing cotton, wood, and silk, can cause eye burns, nausea, vomiting, and mental confusion, sometimes leading to permanent damage.[84] Malachite green, prevalent in the textile industry and as an additive in paints and printing inks, negatively impacts the immune and reproductive systems and damages vital organs such as the spleen, liver, and kidneys.[85] Azo dyes, commonly found in food products like chili powders and palm oils, are highly mutagenic and carcinogenic, often leading to liver and bladder tumors in mammals.[86] Similarly, methyl orange, used in food, plastics, and textiles, is a hazardous water pollutant with mutagenic and carcinogenic properties.[87] Acid red 27, employed in the food and textile industries and color photography, is carcinogenic and toxic to marine life, flora, and fauna.[88,89] Acid black 210, used in cotton, leather, and wool dyeing, causes skin and eye irritation, while acid yellow, applied to wool, silk, and leather, is environmentally harmful.[90,91] Eriochrome black T, used in carpets and textiles, degrades water quality and causes eye and skin allergies.[92] Crystal violet, applied in paper and textile industries and as a temporary hair colorant, contaminates groundwater and is linked to severe health issues, including heartbeat irregularities and tissue necrosis.[93] Congo red, found in leather and paper industries, damages DNA and ecosystems and is associated with neurodegenerative diseases.[94] Finally, reactive blue 19, used in textiles, impacts water solubility due to its high stability and intense coloration, making it persistent in the environment.[95] In most cases, dyes are used to enhance the appearance and quality of products. However, during production and after use, leftover dyes contribute to numerous health and environmental risks. Industries such as pharmaceuticals, wastewater treatment plants, food production, leather processing, households, textiles, and paints commonly utilize dyes in their processes.[77] Fig. 3 illustrates the direct and indirect impacts of dyes on human health and aquatic ecosystems.

## 4. Synthetic methods for preparing CD/MO hybrids

Several methods are commonly employed for synthesizing CD/MO hybrid materials, each offering unique advantages. Hydrothermal synthesis involves heating a mixture of precursors for both carbon dots and metal oxides in separate sealed autoclaves, followed by hydrothermal mixing. This process promotes the *in situ* growth of CDs on metal oxide surfaces or the simultaneous formation of both components, resulting in uniform dispersion and strong interfacial bonding.[96,97] Physical mixing and ultrasonication involve blending pre-synthesized CDs and MOs, typically with ultrasonication or vigorous stirring to enhance dispersion, although this approach may result in weaker interfacial interactions.[98] In the sol–gel method, a metal oxide precursor is hydrolyzed and condensed in the presence of CDs, followed by drying and calcination to obtain the final hybrid material, allowing precise control over composition and particle size.[99] Co-precipitation entails the simultaneous precipitation of metal ions and CDs by adjusting the pH (using agents like NaOH or $NH_4OH$), leading to the deposition of MOs onto the CD surface or *vice versa*; this method is simple and scalable.[100]

## 5. Dye removal methods

In the late 1990s, dye removal primarily relied on basic water purification techniques like equalization and sedimentation, as there were no established limits for dye effluent discharge.[101] With the introduction of permissible discharge standards, more advanced dye removal methods were developed, including dye-degrading filter beds and activated sludge processes.[23] Currently, extensive research is being conducted to identify the optimal dye removal method, enabling the recovery and reuse

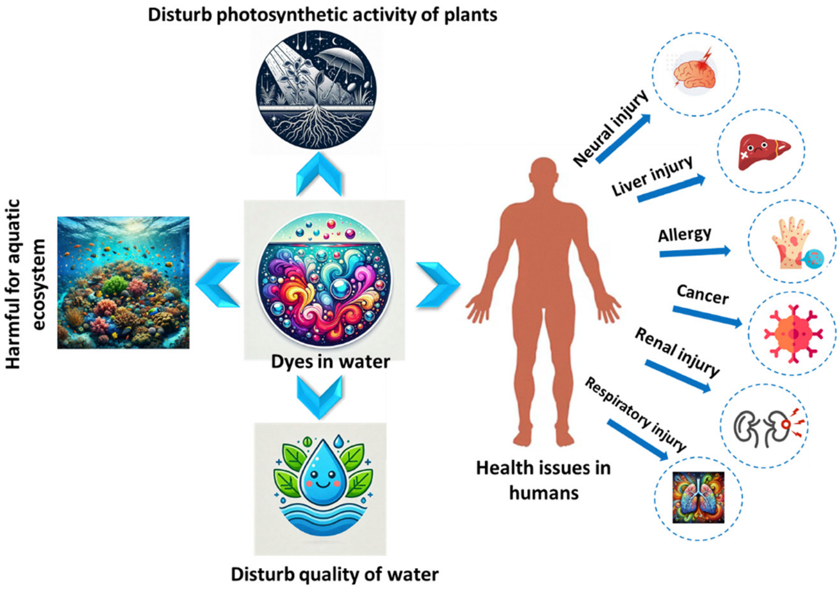

Fig. 3 Impacts of dyes on aquatic ecosystems and human health.

of dye wastewater.[102] Existing dye removal techniques are broadly classified into biological, chemical, and physical treatments.[103] Despite significant advancements in dye removal methods over the past three decades, only a few are widely adopted by industries today due to the limitations associated with most approaches.[104]

### 5.1. Biological dye removal methods

Bioremediation offers a cost-effective solution to address wastewater treatment challenges efficiently. This approach utilizes plants (phytoremediation), algae (phycoremediation), fungi (mycoremediation), and bacteria (biodegradation) to break down organic and inorganic pollutants into less toxic forms.[105] The bioremediation of organic compounds occurs directly at contaminated sites, often using indigenous or native microorganisms, or those isolated from other locations, a process known as bioaugmentation.[106] Using microalgae and bacterial consortia in treating textile wastewater is economical and environmentally friendly. This strategy promotes nutrient recovery and biomass production and supports the bioeconomy. Bacteria enhance microalgae growth in symbiotic consortia, and microalgae reciprocate this effect.[107] Microalgae, with their high surface area and binding affinity, exhibit excellent sorption efficiency and facilitate electrostatic attraction. They can thrive in harsh conditions such as elevated pH and high salinity.[108] Despite its advantages, bioremediation often requires additional nutritional supplements (*e.g.*, carbon and nitrogen sources) or prior acclimatization to compensate for the nutrient-poor nature of dye effluents.[109] The core of this technique lies in the bio-transformation of hazardous waste into simpler, non-toxic compounds. Microorganisms play a crucial role due to their flexibility in degrading diverse compounds. Dye decolorization through bioremediation mainly involves adsorption and degradation occurring in aerobic or anaerobic conditions. Key factors influencing biological methods include the type and concentration of biological species, temperature, pH, and the initial dye concentration.[110] Microorganisms like bacteria, filamentous fungi, algae, yeast, and their enzymes have garnered significant interest for their efficiency, cost-effectiveness, and availability in large quantities. They can degrade a wide range of dyes and convert dye molecules into non-toxic forms.

### 5.2. Chemical dye removal methods

Chemical dye removal methods rely on chemical principles and reactions to eliminate dyes. Common techniques include electrochemical destruction, Fenton reaction, oxidation, advanced oxidation processes, ozonation, photochemical methods, and ultraviolet (UV) irradiation. Various chemical dye removal methods offer advantages and disadvantages based on their processes and applications. Advanced oxidation processes effectively eliminate toxic materials and dyes under unusual conditions but are expensive, pH-dependent, and produce undesirable byproducts.[14] Electrochemical destruction avoids chemical consumption and sludge buildup but incurs high electricity costs and produces hazardous materials with reduced efficiency at high flow rates.[9] The Fenton reaction is effective for soluble and insoluble dyes, removing toxins and accommodating solid content. However, it generates high iron sludge, requires low pH, and has a long reaction time.[111] Oxidation processes degrade dyes into carbon dioxide and water quickly and efficiently, but they are costly, pH-sensitive, and rely on catalysts for optimal results.[112] Ozonation efficiently removes dyes without increasing wastewater volume or generating sludge, yet its high cost, short ozone half-life, and production of toxic byproducts are drawbacks.[113] Photochemical methods combining the Fenton reaction with UV light avoid sludge and foul odors but are expensive and yield numerous byproducts.[114] Similarly, ultraviolet irradiation weakens foul odors and avoids sludge generation but requires hazardous chemicals, consumes significant energy, and has limited treatment capabilities.[115]

### 5.3. Physical dye removal methods

Physical dye removal methods are simple techniques based on mass transfer processes, including adsorption, coagulation or flocculation, irradiation, ion exchange, membrane filtration, and reverse osmosis.[116] Adsorption uses high-capacity adsorbents to trap dye molecules effectively, but can be expensive. Coagulation and flocculation involve adding agents to dye wastewater to form clumps that are filtered out, making it cost-effective yet limited to certain dye types and generating significant sludge. Ion exchange swaps ions in the dye wastewater with those on a solid surface, producing high-quality water but with limited applicability. Irradiation, effective at a laboratory scale, is costly and requires substantial dissolved oxygen. Membrane filtration and its advanced forms, such as nanofiltration and ultrafiltration, effectively recover water but face high costs, frequent fouling, and concentrated sludge production. Reverse osmosis, a pressure-driven system, is widely used for decolorizing and desalting dyes, yielding pure water but at a high cost and requiring substantial pressure.[70,117]

### 5.4. Nanotechnological approaches

Nanotechnology is regarded as an advanced approach for remediating organic pollutants, incorporating the design of safe nanomaterials that foster ecological development and provide environmental benefits.[118] This technology involves manipulating matter on the nanoscale (1–100 nm) across disciplines such as chemistry, engineering, biology, and material science.[119,120] Nanotechnological methods, including nano-adsorbents,[121] carbon nanotubes (CNTs),[122] MOFs,[123] and nanomaterials like titanium dioxide nanoparticles ($TiO_2$ NPs),[124] zinc oxide nanoparticles (ZnO NPs),[125] nanozyme-based approaches,[126] and CDs,[127] demonstrate significant potential for efficient, cost-effective organic pollutant removal.[128] The adsorption capacity of these nanomaterials is largely attributed to their surface properties and porous structures. Nanotechnological approaches based on CDs and MO technologies are discussed in this review.

### 5.5. Photocatalytic degradation mechanism of CD/MO for dye removal

Photocatalytic degradation has emerged as a highly efficient and environmentally benign approach for the removal of synthetic dyes from wastewater. Among various photocatalysts, CD/MO composites have attracted considerable interest due to their superior light-harvesting capability, efficient charge carrier separation, and enhanced photostability under irradiation. The incorporation of CDs into metal oxides such as titanium dioxide ($TiO_2$), zinc oxide (ZnO), and magnetite ($Fe_3O_4$) results in synergistic effects that significantly enhance photocatalytic performance under both UV and visible light. As depicted in Fig. 4.

When the composite material is exposed to a suitable light source specifically, one whose photon energy equals or exceeds the bandgap energy of the semiconductor photocatalyst two fundamental reactions occur concurrently: an oxidation process facilitated by photogenerated holes ($h^+$) and a reduction process driven by photogenerated electrons ($e^-$).[58] Upon irradiation, both CDs and metal oxide nanoparticles absorb photons. CDs, with their tunable bandgap and up-conversion photoluminescence properties, can convert low-energy visible or near-infrared photons into higher-energy emissions capable of exciting metal oxide semiconductors.[129–131] This excitation promotes electrons from the valence band (VB) to the conduction band (CB) of the metal oxide, generating electron–hole pairs, as shown in the equation below:

$$\mathrm{MO} + h\nu \rightarrow \mathrm{e^-(CB)} + \mathrm{h^+(VB)}$$

Farjadfar *et al.* investigated this mechanism using the CD@C-m$TiO_2$ composite and identified the active species responsible for dye degradation by employing radical scavengers. Their findings confirmed that superoxide radicals ($^\bullet O_2^-$), hydroxyl radicals ($^\bullet OH$), and photogenerated holes ($h^+$) are the dominant reactive species contributing to the photocatalytic process. The embedded CDs not only enhanced visible-light absorption but also played a critical role in promoting charge carrier separation, as confirmed by photoluminescence spectra, which showed suppressed electron–hole recombination in the composite. The enhancement in photocatalytic activity can thus be attributed to two key effects: (i) the improved absorption of visible light due to the sensitizing effect of CDs, and (ii) the facilitated separation and transfer of photogenerated charge carriers. The proposed mechanism, illustrated in Fig. 5, shows that upon visible light irradiation, carbonized mesocrystalline $TiO_2$ (C-m$TiO_2$) absorbs light energy, resulting in the excitation of electrons from the VB to the CB. The CDs immediately capture the excited electrons, effectively preventing recombination, while the remaining holes in the VB can oxidize water molecules or hydroxide ions to form hydroxyl radicals, which are highly reactive and responsible for the oxidative degradation of dye molecules.[61]

Furthermore, Zhang *et al.* demonstrated a similar mechanism in CDs/N-$TiO_2$ photocatalysts. Their study emphasized that nitrogen doping narrows the bandgap of $TiO_2$, enabling it to respond more effectively to visible light. This modification facilitates electron transfer from $TiO_2$ to the CDs, thereby improving overall photocatalytic performance.[132]

## 6. Applications

### 6.1. Dye removal using carbon dots with titanium oxide

Doping is a highly effective technique for modifying the optical and electrical properties of bulk semiconductor materials. Research has demonstrated that CDs possess a small band gap and absorb light in the visible spectrum, making them potential candidates for photocatalytic applications.[133]

Titanium dioxide ($TiO_2$) is extensively acknowledged as a significant photocatalyst due to its notable activity under ultraviolet (UV) light. To be deemed an efficient and practical photocatalyst, a material must exhibit the ability to undergo oxidation state changes without experiencing decomposition. $TiO_2$ is distinguished by its low cost compared to alternative materials for water treatment, ease of synthesis in laboratory conditions, exceptional physical and chemical stability, and non-toxic nature, making it a preferred candidate for photocatalytic applications.[134,135] However, the practical application of $TiO_2$ in large-scale photocatalytic processes remains limited

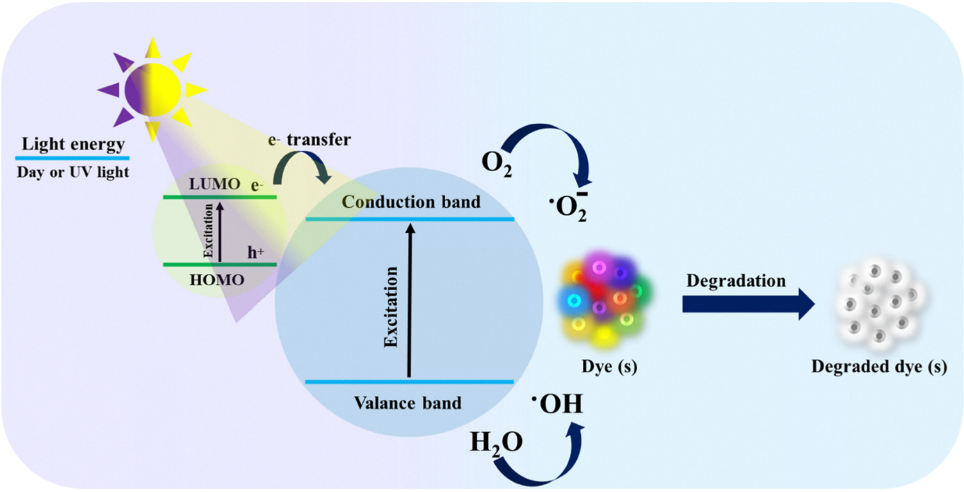

Fig. 4 Schematic representation of the photocatalytic dye removal process by CD/MO hybrid materials.

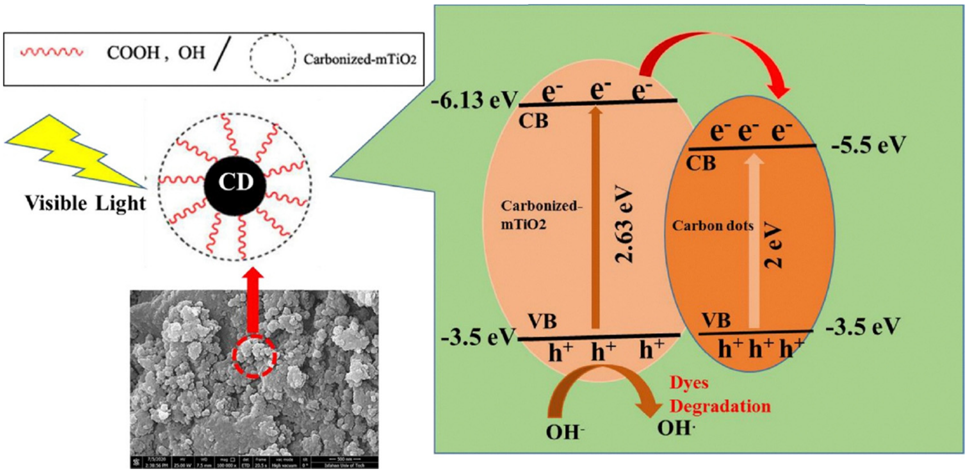

Fig. 5 A schematic illustrating the processes of charge transfer and free radical formation in CD@C-m$TiO_2$ composites that drive their photocatalytic activity. Reproduced with permission from ref. 61. Copyright 2022, Elsevier.

due to its wide band gap and the rapid recombination of photogenerated electron–hole ($e^-/h^+$) pairs, significantly reducing its efficiency. Various strategies, such as metal or non-metal doping and coupling with small band gap semiconductors, have been developed to address these limitations to enhance its photocatalytic performance.[136] Additionally, recent research has explored coating $TiO_2$ with CDs as an innovative approach to address the recombination of electron–hole pairs. This method has been shown to enhance energy absorption by $TiO_2$, facilitate electronic transitions, increase the generation of electron–hole pairs, and produce reactive oxygen species under light irradiation. As a result, highly efficient photocatalysts have been developed.[137,138] Jin *et al.*[139] reported using N-doped carbon quantum dots (NCQDs) to decorate $TiO_2$ through a straightforward hydrothermal-calcination synthesis method. The NCQDs enhanced visible light absorption and promoted efficient electron transfer while improving the separation of photogenerated electron–hole pairs. With just 3 wt% NCQDs (3-NCQDs/$TiO_2$), the photodegradation rate of methylene blue (MB) was accelerated by 2.25 times compared to pure $TiO_2$. The 3-NCQDs/$TiO_2$ catalyst demonstrated excellent and stable photocatalytic activity, achieving a 93.1% photodegradation rate of MB within 60 minutes and maintaining over 86% efficiency after four cycles. These results were obtained using a catalyst dose of 100 mg and an initial MB concentration of 10 ppm.

Nu *et al.*[140] synthesized carbon dots derived from microalgae (MCDs) and successfully integrated them onto the surface of $TiO_2$ nanoparticles. The resulting $TiO_2$-MCD composite demonstrated significantly enhanced photodegradation efficiency for MB under visible light irradiation at room temperature. Specifically, the composite achieved an 83% degradation rate compared to 27% for pristine $TiO_2$, with an initial MB concentration of 10 ppm. In this system, the MCDs act as electron reservoirs, trapping photoinduced electrons and as photosensitizers to enhance visible light absorption. These roles collectively contribute to the improved photocatalytic performance of the $TiO_2$-MCD composite, as illustrated in Fig. 6.

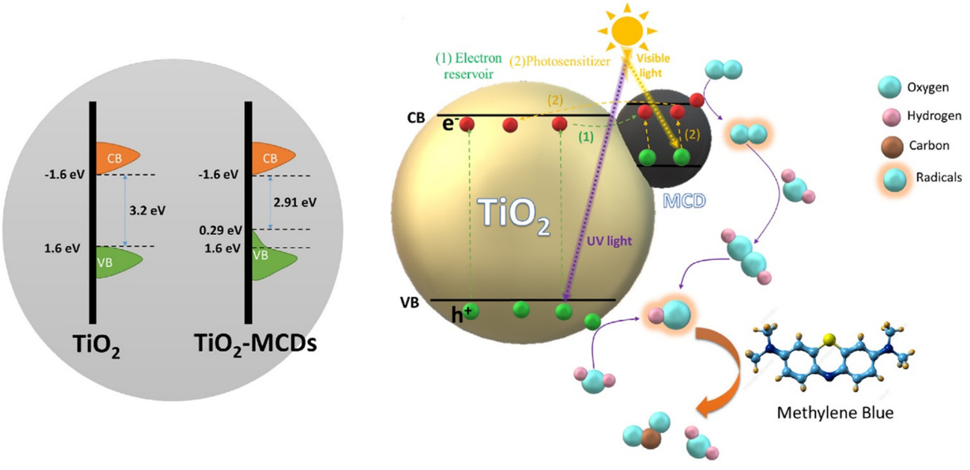

Fig. 6 A schematic depiction of the photocatalytic mechanism demonstrated by $TiO_2$-MCDs. Reproduced with permission from ref. 140. Copyright 2022, Elsevier.

Heng *et al.*[141] proposed utilizing waste oil palm fronds as a sustainable precursor for NCQDs to synthesize NCQDs/$TiO_2$ nanocomposites. For the first time, NCQDs/$TiO_2$ derived from oil palm fronds were employed in the photodegradation of MB under visible light. The nanocomposite exhibited a photocatalytic efficiency of 86.16%, 2.85 times higher than that of $TiO_2$ alone (30.18%). This improved performance was attributed to NCQDs acting as an electron mediator and visible-light harvester. The optimal NCQD loading was 4 mL, with the highest removal efficiency achieved at a catalyst dosage of 1 g $L^{-1}$ under alkaline conditions. The initial dye concentration was 10 mg $L^{-1}$.

Table 1 summarizes studies that utilized CDs/$TiO_2$ hybrid material for the removal of multiple dyes.

### 6.2. Dye removal using carbon dots with zinc oxides

Zinc oxide (ZnO) is considered a promising photocatalyst due to its high catalytic activity, low cost, wide bandgap (3.37 eV), and environmental friendliness.[162] However, its rapid recombination of charge carriers and its narrow absorption band significantly affect its photocatalytic efficiency. To address these challenges, appropriate modifications are necessary to develop advanced ZnO nanostructures. This can be achieved by incorporating either secondary semiconductor metals[163] or carbon nanoparticles.[164] CDs, for instance, act as electron reservoirs, enhancing the separation efficiency of photogenerated charge carriers and boosting photocatalytic performance.[165] Coating ZnO nanoparticles (NPs) onto CDs is also promising to improve charge separation, prevent photo-corrosion, and enhance overall photocatalytic efficiency.[166] Kumar *et al.*[167] investigated the formation of heterojunctions composed of GQDs decorated on ZnO and their application as efficient photocatalysts for environmental remediation. The study focused on the photocatalytic performance of these heterojunctions in degrading the colored pollutant MB dye. Among the tested configurations, the heterojunction with 2 wt% GQD (ZGQD) exhibited the highest photocatalytic activity, achieving approximately 95% degradation of MB. The enhanced photocatalytic efficiency of the ZnO-GQD heterojunctions can be attributed to the efficient charge carrier separation, which suppresses the recombination rate at the photocatalyst interfaces. Additionally, the improved light absorption from the UV to the visible spectrum, combined with the high specific surface area (353.447 $m^2$ $g^{-1}$), contributes to its strong pollutant adsorption capacity on the catalyst surface, enhancing its photoactivity.

Sekar *et al.*[168] investigated using CZnO-dots, a visible-light-responsive photocatalyst, to decolorize MG dye efficiently. The optical bandgap was determined using a Tauc plot, revealing that combining ZnO with CDs reduces the bandgap compared to bare CDs and pure ZnO nanoparticles. CZnO-dots with 1 wt% ZnO exhibited the highest photocatalytic efficacy among the tested samples. Under optimized conditions, the CZnO-dots achieved a degradation efficiency of approximately 94.8% within 60 minutes of irradiation. The study also explored the mechanism behind MG dye's photocatalytic degradation, including identifying active species in the photodegraded products. Furthermore, an environmental assessment was conducted to evaluate the potential of treated dye water for developing mung bean plants. Experimental results showed effective performance using a catalyst dose of 1.5 mg of CZnO-dots with an initial dye concentration of 0.05 g $L^{-1}$, as depicted in Fig. 7.

Table 2 summarizes studies that utilized the CDs/ZnO hybrid material to remove multiple dyes.

### 6.3. Dye removal using carbon dots with iron oxides

Magnetite ($Fe_3O_4$) and maghemite ($\gamma$-$Fe_2O_3$) are the most common iron oxide phases, characterized by exceptional physicochemical properties such as high surface area, environmental

Table 1 Removal of dyes using a CDs/$TiO_2$ hybrid

| Method | Dye | Catalyst dosage | Initial concentration | Performance % | Ref. |
|---|---|---|---|---|---|
| Degradation | MB | 15 mg | 10 mg $L^{-1}$ | 90 | 142 |
| Degradation | MB | 0.1 g | 10 mg $L^{-1}$ | 86 | 143 |
| Degradation | ST | — | — | — | 144 |
| Degradation | MB | 25 mg | 10 ppm | 75 | 145 |
| Degradation | MO | 50 mg | 10 mg $L^{-1}$ | 95.5 | 146 |
| Degradation | MB | 20 mg | 10 mg $L^{-1}$ | 85 | 147 |
| Degradation | MO | 20 mg | 10 ppm | 79 | 148 |
| Degradation | MB | 20 mg | — | 90 | 149 |
| Degradation | RhB | 30 mg | 10 mg $L^{-1}$ | 99.8 | 132 |
| Degradation | MB | 0.2 g | 10 ppm | 85 | 133 |
| Degradation | MB | 0.1 | 10 mg $L^{-1}$ | 86.9 | 150 |
| Degradation | RhB | 50 mg | 5 ppm | 100 | 151 |
| Degradation | RhB | 1 mg $mL^{-1}$ | — | 99 | 136 |
| Degradation | MB | 50 mg | 10 μg $mL^{-1}$ | 90 | 152 |
| Degradation | AO | 50 mg | 10 mg $L^{-1}$ | — | 153 |
| Degradation | RhB | 0.1 g | $1 \times 10^{-5}$ mol $L^{-1}$ | 95 | 154 |
| Degradation | Direct red 23, direct red 80 | — | — | 60, 50 | 60 |
| Degradation | MO | 0.02 g | 10 mg $L^{-1}$ | 60 | 155 |
| Degradation | MO | 50 mg | 5 mg $L^{-1}$ | 95 | 156 |
| Degradation | RhB | 15 mg | 10.0 mg $L^{-1}$ | 96 | 157 |
| Degradation | MB | 20 mg | 10 mg $L^{-1}$ | 99.23 | 158 |
| Degradation | MB | 1 g | 20 mg $L^{-1}$ | — | 159 |
| Degradation | RhB | 10 mg | 20.0 mg $L^{-1}$ | 94, 37 | 160 |
| Degradation | Reactive black 5 | 0.8 g $L^{-1}$ | 50 ppm | 100 | 161 |

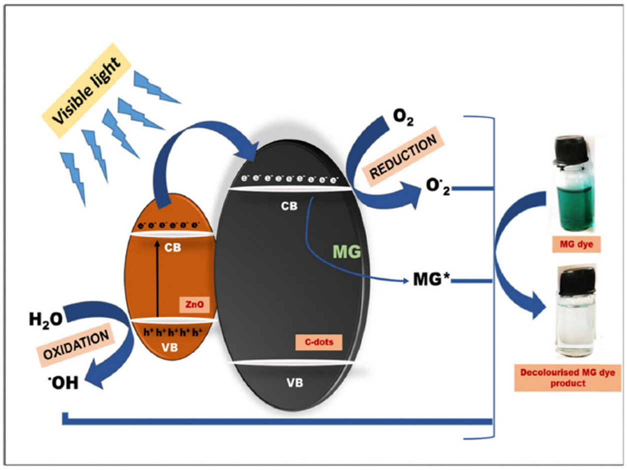

Fig. 7 Proposed mechanism for the photodegradation of MG using CZnO-dots. Reproduced with permission from ref. 168. Copyright 2021, Elsevier.

compatibility, superparamagnetic behavior, non-toxicity, and cost-effectiveness. $Fe_2O_3$ has garnered significant interest among transition metal oxides due to its remarkable attributes.[183] CDs, as a novel class of nanocarbon materials, have gained widespread attention for their strong fluorescence, superior conductivity, and rapid electron transfer capabilities. Notably, the abundant functional groups on the surface of CDs serve as effective sites for immobilizing metal nanoparticles, thereby enhancing the stability and catalytic activity of the parent materials.[184] In particular, incorporating CDs has emerged as a promising strategy for improving the photocatalytic performance of iron oxide-based materials.[185]

Sajjadi *et al.*[186] synthesized $Fe_3O_4$ nanoparticles functionalized with nitrogen- and sulfur-co-doped graphene quantum dots (NS-GQDs) and evaluated their performance as a sonocatalyst. The NS-GQD-decorated $Fe_3O_4$ (NS-GQD/$Fe_3O_4$) nanoparticles exhibited a significantly enhanced decolorization efficiency of 99.0%, compared to 43.7% achieved with pure $Fe_3O_4$ nanoparticles. The bandgap of the NS-GQD/$Fe_3O_4$ nanostructures (1.923 eV) was notably lower than that of pure $Fe_3O_4$ nanoparticles (2.147 eV). Optimal catalytic performance was obtained with a dopant concentration of 3 wt%. The highest sonocatalytic decolorization efficiency of 99% was observed at an initial pH of 8. In this study, the initial MB concentration was 20 mg L$^{-1}$, and the catalyst dosage was maintained at 1 g L$^{-1}$. Similarly, Chen *et al.*[187] successfully synthesized a $Fe_3O_4$@CQDs magnetic nanocatalyst by modifying α-$Fe_2O_3$ and demonstrated its efficacy in degrading rhodamine B (RhB) through the activation of peroxymonosulfate (PMS) under visible light. To investigate the superior activation performance of the $Fe_3O_4$@CQDs compared to α-$Fe_2O_3$ at the microscopic level and elucidate the reaction mechanism, density functional theory (DFT) calculations were conducted. The results indicated that the RhB degradation efficiency reached 98% within 35 minutes. The experimental conditions included a catalyst dosage of 50 mg and an initial RhB concentration of 20 mg L$^{-1}$.

Table 3 Removal of dyes using a CDs/iron oxide hybrid

| Method | Dye | Catalyst dosage | Initial concentration | Performance % | Ref. |
|---|---|---|---|---|---|
| Degradation | MB | 20 mg | 20 mg L$^{-1}$ | 84 | 188 |
| Degradation | MB | 0.05 g | 20 mg L$^{-1}$ | 85.6 | 188 |
| Degradation | RhB | 50 mg | 10 mg L$^{-1}$ | 90 | 189 |
| Degradation | CV | 0.1 mg mL$^{-1}$ | 0.05 mM | — | 185 |
| Degradation | RhB | 0.03 g | 10 ppm | 92 | 190 |

Table 3 summarizes studies that utilized the CDs/iron oxide hybrid material to remove multiple dyes.

Various metal oxide/carbon dot hybrids have been synthesized and employed to remove multiple dye contaminants. These hybrids incorporate metal oxides such as copper oxide (CuO), cerium oxide ($CeO_2$), manganese oxide ($MnO_x$), *etc.*, each selected for its unique properties that enhance the adsorption and degradation of dyes. Table 4 comprehensively summarizes these hybrid materials, detailing their synthesis methods, structural characteristics, and efficiencies in removing various dyes. The synergistic interactions between the metal oxides and carbon dots in these composites contribute to improved catalytic activity, increased surface area, and enhanced electron transfer capabilities, making them highly effective for wastewater treatment applications.

Table 2 Removal of dyes using a CDs/ZnO hybrid

| Method | Dye | Catalyst dosage | Initial concentration | Performance % | Ref. |
|---|---|---|---|---|---|
| Degradation | MG | 5 mg | $1.0 \times 10^{-4}$ M | 82 | 169 |
| Degradation | MO | 40 mg | 10 mg L$^{-1}$ | 91.25 | 170 |
| Degradation | MO, MB, RhB | 50 mg L$^{-1}$ | 10 mg L$^{-1}$ | 91, 99, 99 | 171 |
| Degradation | MB | — | 10 mM | 81.13 | 172 |
| Degradation | MG | 0.05 g L$^{-1}$ | 20 mg L$^{-1}$ | 85 | 173 |
| Degradation | MB | 20 mg | — | 95 | 174 |
| Degradation | MB | 100 mg L$^{-1}$ | 10 mg L$^{-1}$ | 96 | 175 |
| Degradation | RO, RR, RB | 20 mg | $5 \times 10^{-5}$ M | 68, 34, 28 | 176 |
| Degradation | MB | 5 mg | 10 ppm | 83.4 | 177 |
| Degradation | MB, MO, RhB | 0.01 g | 5 mg mL$^{-1}$ | 90.87%, 57.95%, 86.70 | 178 |
| Degradation | MB | 20 mg | — | 99 | 179 |
| Degradation | RhB | 50 mg | 10 ppm | 92 | 180 |
| Degradation | RhB | 30 mg | $1 \times 10^{-5}$ M | 98.7 | 181 |
| Degradation | MB | 0.03 g | 5 ppm | 98.17 | 182 |

Table 4 Removal of dyes using a CDs/metal oxide hybrid

| Method | Type of MOs | Dye | Catalyst dosage | Initial concentration | Performance % | Ref. |
|---|---|---|---|---|---|---|
| Degradation | $SnO_2$ | RhB | 80 mg | 10 mg $L^{-1}$ | 95 | 191 |
| Degradation | CuO | MO | 50 mg | 20 ppm | 92 | 192 |
| Degradation | $Ag_3PO_4$ | MB, RhB | 30 mg | 10 mg $L^{-1}$ | 99 | 193 |
| Degradation | $MnO_2$ | RhB | | 10 g $L^{-1}$ | 96.39 | 194 |
| Degradation | $ZrO_2$/CaO/MgO | MB | | 10 ppm | 99.54 | 195 |
| Degradation | $CeO_x$ | MB | 50 mg | | | 196 |
| Degradation | $BiVO_4$ | MB | 50 mg | 10 mg $L^{-1}$ | 90 | 197 |
| Degradation | $CeO_2$ | RhB | 25 mg | 10 mg $L^{-1}$ | 96.9 | 198 |
| Degradation | $CeZrO_2$@$Bi_2MoO_6$ | RhB | 0.05 g | 10 mg $L^{-1}$ | 100 | 199 |
| Degradation | $Bi_2O_4$ | MO | 0.05 g | 10 mg $L^{-1}$ | — | 200 |
| Degradation | $Bi_2MoO_6$ | RhB, MB | 80 mg | 10 mg $L^{-1}$ | 99.4, 75 | 201 |
| Degradation | $BiVO_4$ | RhB | 20 mg | 10 mg $L^{-1}$ | 98 | 202 |
| Degradation | CaO | RhB | | | 77.83 | 203 |
| Degradation | $Ag_3PO_4$ | Neutral red | 50 mg | 12 mg $L^{-1}$ | 95.5 | 204 |
| Degradation | $KNbO_3$ | CV | 50 mg | 10 mg $L^{-1}$ | 70 | 205 |
| Degradation | MgO | MB | 0.003 g | 5 mg $L^{-1}$ | 99.9 | 206 |
| Degradation | $WO_3$ | RhB, MB | 50 mg | 10 mg $L^{-1}$ | 97.1, 91.1 | 207 |
| Degradation | $CuCo_2O_4$ | MB | 5 mg | 5 mg $L^{-1}$ | 96.4 | 208 |
| Degradation | $Cu_2O$ | MB. RhB | 50 mg | 10 mg $L^{-1}$, 5 mg $L^{-1}$ | 100 | 209 |
| Degradation | $La_2O_3$ | RhB | — | — | 94.57 | 210 |
| Degradation | $Cu_2O$ | MB | — | 10 mg $L^{-1}$ | 96.4 | 211 |
| Degradation | $MnCo_2O_4$ | AB, CV, and MB | 50 mg | 20 ppm | 94, 82.33, 67.5 | 212 |
| Degradation | $MnO_2$ | MG, MV | 1 mg | 10 ppm | 96.5, 96 | 213 |
| Degradation | BiOI | MB | 50 mg | $1.5 \times 10^{-5}$ mol $L^{-1}$ | 76.2 | 214 |

## 7. Conclusions and future prospects

Since the discovery of CDs in 2004, their application in photocatalysis has garnered significant attention in the research community, primarily due to their ability to bind with various bulk metal oxides for effective dye removal. This review summarizes design strategies for CD-based metal oxide photocatalysts, evaluates recent advancements, and assesses their photocatalytic performance in degrading a range of dyes. It also discusses the types of dyes present in water and their associated toxicity, highlighting these pollutants' environmental and health impacts. Consequently, there is a pressing need to eliminate or reduce dye contamination in water sources. CDs combined with metal oxides are hybrid materials for dye removal, leveraging their robust properties. The synergistic effects arising from this combination often surpass the performance of the individual components. Previous studies indicate that several technical factors, including the tuning of the band structures and the composites' optical properties, largely influence the efficiency of photocatalytic systems. Key parameters that can be optimized include surface morphology, crystal structure and exposed facets, band gap, and the position of the band edges of the metal oxides, as well as surface doping and the composition of CDs in the nanocomposite.

Despite the intelligent design of CD-based photocatalysts and their enhanced catalytic performance, several challenges in this field warrant further exploration. Notably, while CDs have various roles in enhancing the photocatalytic activity of metal oxide systems, the direct application of bare CDs as photocatalysts is infrequently studied and underutilized. Recent research indicates that CDs possess significant potential as standalone photocatalysts due to their narrow band gap between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO). Therefore, more fundamental studies and in-depth analyses of the photocatalytic capabilities of bare CDs are necessary to realize their full potential in photocatalytic reactions.

The removal of toxic dyes from wastewater using CD/metal oxide hybrid materials presents both significant challenges and promising opportunities. Dyes are structurally complex, highly stable, and often toxic, posing serious risks to aquatic ecosystems and human health even at low concentrations. While CD/metal oxide hybrids offer enhanced photocatalytic and adsorptive properties, issues such as limited stability, poor reusability, interference from coexisting pollutants, and difficulties in achieving efficient synergy between components hinder their practical application. Additionally, the scalability and cost-effectiveness of current synthesis methods remain major barriers. Nevertheless, future prospects are encouraging, including the design of surface-functionalized and doped CDs to optimize charge transfer, the development of green and scalable synthesis routes, and the incorporation of multifunctionality (*e.g.*, magnetic recovery or membrane integration) to improve real-world applicability. A deeper mechanistic understanding and comprehensive environmental risk assessment are also essential to ensure the safe and effective deployment of these materials in wastewater treatment systems.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

Data sharing is not applicable to this article as no datasets were generated or analysed during the current study.

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