



Removal of Dyes Used Pharmaceutical and Other Industries from Wastewater Using Metal Oxides and Carbon Dots as Hybrid Materials: A review

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Abstract

Many modern technologies based on luminescent materials play important roles in visualization devices, lighting systems, X-ray imaging, and scintillation applications. This review condenses the latest developments in near-infrared (NIR) emission produced by rare-earth and transition-metal activation and describes the applications of these emissions in the fields of optoelectronics, bioimaging, and sensing. Researchers have been looking for new and better luminescent systems as these applications grow. Defect-based luminescent materials have gained a lot of attention in the past ten years. These materials typically fall into a number of categories, such as carbon-based emitters, BCNO phosphor, metal oxides, and hosts derived from silica. One of the main benefits of defect-related phosphors is that they can be produced in large quantities and at a low-cost using methods like chemical vapor deposition, hydrothermal synthesis, sol-gel processing, and sonochemical approaches. In this review, we provide an overview of recent advancements in their preparation techniques, emission behavior, and optical property tuning and control strategies. Lastly, we discuss potential future paths and new prospects for applying these materials to biomedical and lighting technologies.

Keywords: Luminescent materials, rare earth and transition metal ions, synthesis, bioimaging, near-infrared luminescence

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1. Introduction

Aquatic pollution is primarily caused by industrial waste, which threatens human health and natural systems at significant risk [1]. Specifically, waste products from the pharmaceutical and textile industry's printing and dyeing processes [2]. It is considered as one of the most severely contaminated forms of industrial waste due to its high levels of chemical oxygen demand (COD), colored pollutants, and hazardous materials [3]. It could produce damaging and unexpected effects on the environment if discharged untreated [4]. Approximately 6 to 8 million individuals die each year as a consequence of water-related illnesses and crises [5]. Therefore, it becomes essential to treat this wastewater to a safe level. To tackle this problem, a number of therapy approaches have been established including biodegradation, electrochemical, chemical precipitation, membrane filtration, coagulation, and adsorption [6]. Within these processes, adsorption and deterioration are frequently chosen for their affordability, simplicity of use, minimal waste, and environmental sustainability [7]. Toxic dyes can be successfully eliminated using a variety of substances, such as organic compounds, metal-organic frameworks (MOFs),

activated carbon, zeolites, sulfur dots, and synthetic polymers [8]. Nevertheless, since they have fewer available binding sites, metallic materials frequently show reduced adsorption ability. Although materials like COFs, MOFs, and synthetic polymers have extremely high capacity to absorb water, their usage is limited by the expensive rare ingredients and difficult production processes [9]. In recent years, research has revolved primarily on CDs, a unique class of carbon-based nanomaterials [10]. In 2004, when purifying single-walled nanotubes made of carbon, these quasi-spherical, zero-dimensional nanoparticles were randomly found [11]. Various CDs with different sizes, designs and luminescent characteristics have been created, carbon quantum dots, polymer dots, and graphene quantum dots are three main types of CDs [12]. The superior photoluminescent characteristics, variety of functional groups, quantity of raw ingredients, simplicity of adaptation, and simple production techniques of CDs differentiate them from other carbon materials [13]. Furthermore, CDs great water absorption, low harmful effects, outstanding biological stability, and remarkable sensitivity to photo bleaching make them suitable substitutes for conventional semiconductor quantum dots

[14]. Due to these advantages, CDs have drawn attention for its diversity, including as drug delivery, bioimaging, sensing, catalysis, and energy storage [15].

Metal oxides, including chromium, vanadium, zinc, tin, cerium, and titanium, have comparable photocatalytic reactions because of their similar characteristics. One of those processes is absorption of light, which causes separation of charges and creates positive ions that can oxidize biological substrates [16]. Whenever a metal oxide is exposed to UV rays, visible wavelengths, or a combination of the two, the charged particles are excited by absorbed energy and move from valence band to conduction band, forming an electron-hole pair (e/h^+). Two processes are mainly responsible for the photocatalytic activity of MOs: (i) the oxidation of hydroxide (OH) anions produces hydroxyl (OH) radicals, and (ii) reduction of molecular oxygen (O_2) produces superoxide (O_2^-) radicals, by interacting with contaminants, these reactive organisms help them degrade or change into less dangerous byproducts [17]. Therefore, introducing CDs to metal oxides is an advantageous means to alter and improve their special electrical, catalytic, and optical characteristics in order to increase their potential uses. Utility and adaptability of metal oxides are significantly increased by the addition of CDs [18]. The development of CDs combined with MOs as well as their role in eliminating harmful dyes are examined in detail for first time in this review. The categorization of dyes, their impact on the human health and environment, dye extraction techniques, and variables influencing dye degradation are all covered. The analysis concludes by examining prospects and difficulties of developing CD/MO hybrid materials for effective dye removal in the future.

1.1. Motivation of this study

Promoting a sustainable ecology will guarantee a healthier environment in the near future. The health of people, aquatic life, terrestrial species, and plants are all negatively impacted by industrial discharges, which also greatly contribute to environmental contamination. Among these pollutants, dye industry waste products distinguish as significant contributors of environmental damage. We were inspired and motivated to conduct this review research by the emerging concern. This assessment emphasizes the possibilities of combining metal oxides and carbon dots as innovative methods for eliminating colors from wastewater to support environmental sustainability. The key objectives of the studies include defining different dyes, comprehend the toxicity of water pollution, and employ photocatalysts and adsorbents based on carbon dots and metal oxides to address this problem. This study aims to contribute to the development of the environment friendly methods for reducing dye pollution and safeguarding the environment by investigating these contemporary alternatives.

2. Dyes and classification

The ecology is being threatened by contaminated water, resulting in severe consequences. Environmental contamination is caused by a variety of impurities, such as metal ions, pesticides, dyes, chemicals, and inorganic substances. The stable aromatic rings found in the majority of dye compounds are the main cause of the persistent and dangerous nature of dyes. An environment may become contaminated by even little levels of color [19]. Because of the adverse impacts on surface and groundwater, wastewater

must be cleaned up before being released [20]. As a result, one of the most hazardous contaminants that has to be tackled is dye pollution. The majority of dyes are not biodegradable since they are resistant to oxidation, light, and heat. Additionally, dyes have an adverse impact on aquatic ecosystems by making natural environments less aesthetically pleasing [21]. A significant group of organic macromolecules, dyes are widely used in numerous fields and have a big impact on our daily lives. Dyes are essential to many industries, including, paints, textiles, metal extraction, dye-sensitized solar cells, optics, sensors and plastics. According to the color index (CI), dyes are categorized according to a number of factors, such as their molecular structure, color, origin and application techniques [22]. Dye molecules comprised of chromophores, that provide dye, and auxochromes, which not only serve to assist chromophore as well as make the molecule more soluble in water and more likely to attach to fibers. Dyes can be classified in a variety of ways and have a wide range of structural variations (Figure 1) [23]. Therefore, few dyes have been categorized with their examples and properties in Table 1.

3. Dyes effects on the human health and environment

Whether they originate from natural or artificial sources, colors are a part of our daily lives and may be seen in the clothing we wear and the items around us. Before the 1800s, minerals were sole provider of color. The main contributions were plants, trees, and lichens; insects and molluscs came in second. Only a small number of these organic dye suppliers regularly employed despite thousands of years of dye consumption, highlighting their volatility. There are currently more than 7000 colorants available on the commercial market [24]. As reported by Samsami et al. (2020), 54% of dye discharges into environment come from textile sector, making it biggest source. Dyeing industries (21%), paint and tannery facilities (8%), paper and pulp factories (10%), and dye production plants (7%) are further significant suppliers. During the manufacture of textiles, several dye combinations are produced [25]. Among most commonly used artificial colors, textile dyes are employed in a variety of sectors, including food, cosmetics, textiles, and pharmaceuticals [26]. By preventing the absorption of solar radiation, which is necessary for photosynthesis and growth of aquatic plants, direct discharge of such dangerous organic waste into surface water bodies disturbs aquatic ecosystems, affecting food chain and creating ecological discrepancies [27]. Furthermore, dyes have been found to be teratogenic, mutagenic, and carcinogenic to a variety of fish species and microorganisms. Dye exposure in humans can cause serious health problems, such as problems with kidneys, liver, brain, reproductive system, and central nervous system [28-29].

Azo dyes are extremely resilient to decomposition and maintain their color in wastewater for long periods of time, but anthraquinone-derived dyes are especially dangerous due to the poisonous amines in their wastewater streams [30]. Around five to ten percent of the volume of reactive water-soluble dyes are released as brightly colored wastewater, which causes serious environmental issues [31]. Although industrial dyes commonly used in many different industries, they constitute serious threats to both environment and human health. Whenever employed in coloring cotton, silk, and wood, methylene blue may result in nausea, vomiting, eye burns, and cognitive impairment, which can

occasionally cause irreversible damage [32]. Malachite green, which is widely used in the textile manufacturing as well as ingredient in the printing inks and paints, affects important organs like liver, spleen, and kidneys and has detrimental effect on immunological and reproductive systems [33]. Eriochrome black T (EBT), a dye which is employed in textiles and carpets, damages water quality and results in allergies to skin and eyes [34]. Congo red, which used in paper and leather fields, harms ecosystems and DNA and linked to neurological disorders [35]. Dyes are typically used to improve product's quality and appearance. However, residual dyes pose a number of health and environmental concerns both during production and after consumption. Dyes are often used in a variety of industries, including medicines, treatment of wastewater, preparing food, textile processing, homes, paints, and textiles (Figure 2) [36].

4. Synthetic methods for CD/MO hybrids

During the synthesis of CD/MO hybrid materials, a number of techniques are often used, each with unique benefits. In hydrothermal synthesis, a combination of precursors for metal oxides and carbon dots is heated in individual sealed autoclaves before being hydrothermally mixed. This approach encourages the *in situ* synthesis of CDs on metal oxide interfaces or the instantaneous production of both constituents, in order to achieve consistent distribution and adhesive attachment [37]. Sol-gel process provides exact control over structure and particle dimension by hydrolyzing and condensing a metal oxide precursor in the proximity of CDs, then air-drying and heating resulting composite material [38]. Co-precipitation is a simple and reproducible process that involves sequentially precipitating metallic ions or CDs by altering pH (with agents like NH_4OH), resulting in the accumulation of MOs on CD surface [39].

5. Dyes removal methods

5.1. Biological dye removal techniques

Biological remediation provides an economical way to effectively handle waste water treatment issues. This method breaks down inorganic as well as organic contaminants into less harmful forms using, algae (phycoremediation), plants (phytoremediation), bacteria (biodegradation), and fungus (mycoremediation) [40]. This approach enhances the bioeconomy by encouraging biomass production and nutrient recovery. In symbiotic partnerships, bacteria promote the development of microalgae, and microalgae [41]. Microalgae display outstanding sorption efficiency and promote electrostatic attraction due to their large surface area and binding affinity. They can survive in challenging environments like high salinity and pH [42]. Although advantages it provides, bioremediation frequently need previous adaptation or supplementary nutritional products (such as carbon and nitrogen sources) to make up for the nutritionally deficient character of dye waste products [43]. The biological transformation of dangerous waste into less complex, harmless molecules is the fundamental component of this method. Adsorption and degradation that takes place in aerobic or anaerobic environments are the primary processes involved in dye decolorization by bioremediation. The kind and number of biological species, pH, temperature, and the starting dye quantity are important variables affecting biological techniques [44].

5.2. Physical dye removal techniques

Physical dye removal methods, such as adsorption, coagulation or flocculation, ion exchange, and reverse osmosis, are straightforward procedures based on mass movement mechanisms [45]. Although the adsorption can be costly, yet it successfully traps color molecules using high-capacity adsorbents [46]. In coagulation and flocculation, additives are incorporated into dye wastewater to generate aggregates that are screened out. This process is economical but only works with specific dyes and produces an immense quantity of sludge. Ion exchange creates high-quality water with minimum consumption by exchanging the ions in the dye waste water with those on a solid surface. Reverse osmosis is a strength-driven technique that is frequently used to desalinate and decolorize dyes. It produces clean water, but it is expensive and requires a lot of force [47].

5.3. Chemical dye removal techniques

Chemical ideas and processes are the basis for chemical dye removal techniques. Electrochemical degradation, oxidation, Fenton reaction, ultraviolet irradiation, advanced oxidation processes, and ozonation are common procedures. Depending on their procedures and uses, several chemical dye removal techniques have advantages as well as limitations. Although they are costly, pH-dependent, and result in unwanted byproducts, advanced oxidation processes successfully remove hazardous chemicals and dyes under exceptional circumstances [48]. Although electrochemical degradation eliminates the need for chemicals and sludge accumulation, it creates hazardous materials at high flow rates with decreased efficiency and high power costs [49]. Toxic substances are eliminated by the Fenton reaction, that performs efficiently for both soluble and insoluble dyes. Nevertheless, it exhibits a prolonged reaction time, produces high iron sludge, and needs a low pH [50]. Oxidation techniques swiftly and effectively break down dyes into water and carbon dioxide, while they are expensive, sensitive to pH, and require additives for optimum results [51]. In a comparable way, UV radiation reduces unpleasant odors and prevents the formation of sludge, but it involves dangerous chemicals, uses a lot of energy, and has limited therapeutic potential [52].

5.4. Nanotechnological methods

In recent years, nanotechnology has become more popular, making water purification more economical and effective [53]. Nanotechnology, that involves the manufacturing of sustainable nanomaterials that promote ecological growth and offer environmental benefits, is thought to be an innovative approach for eliminating organic contaminants [54]. Across the fields including chemistry, engineering, biology, and material science, this technique manipulates matter at the nanoscale (1–100 nm) [55]. Nanotechnological techniques, such as carbon nanotubes (CNTs), MOFs, titanium dioxide nanoparticles, zinc oxide nanoparticles, nanozyme-based methods, CDs, and nanoadsorbents, exhibit enormous potential for effective and economical removal of organic pollutants (Figure 3) [56].

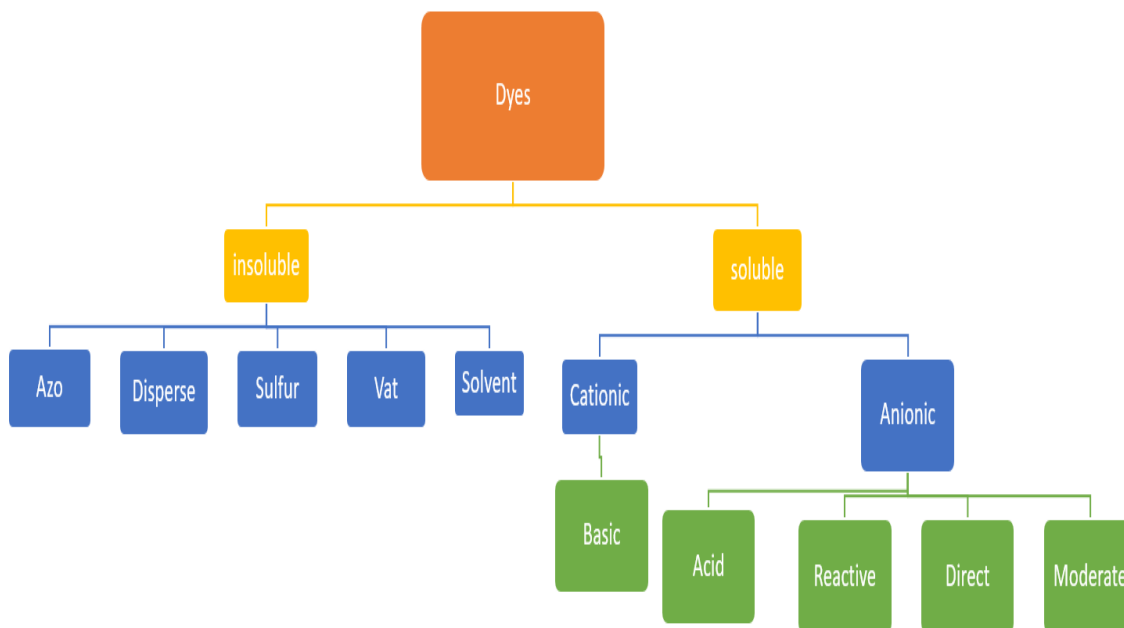


Figure 4. Classification of dyes based on their solubility

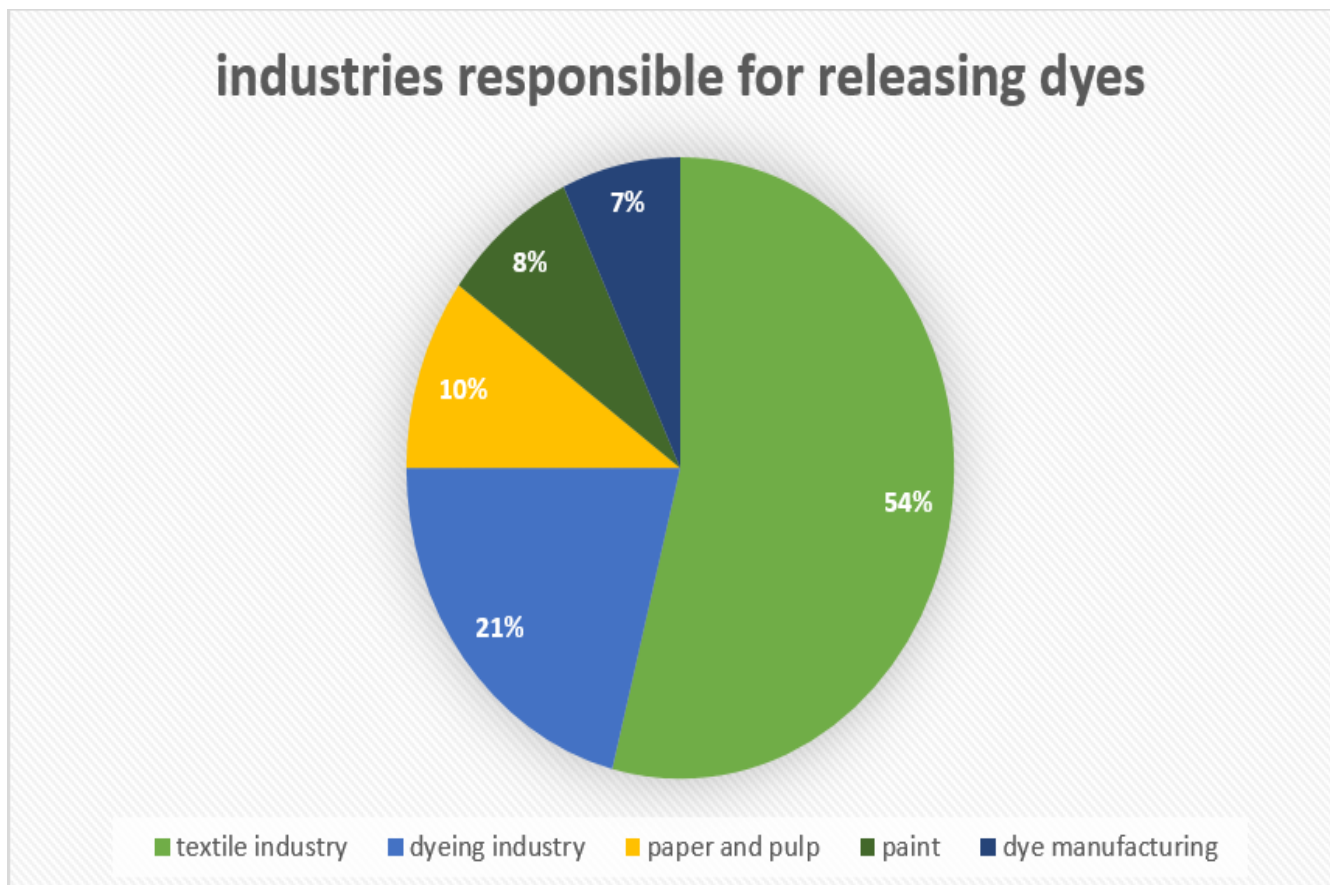


Figure 5. Industries responsible for releasing dyes into the environment

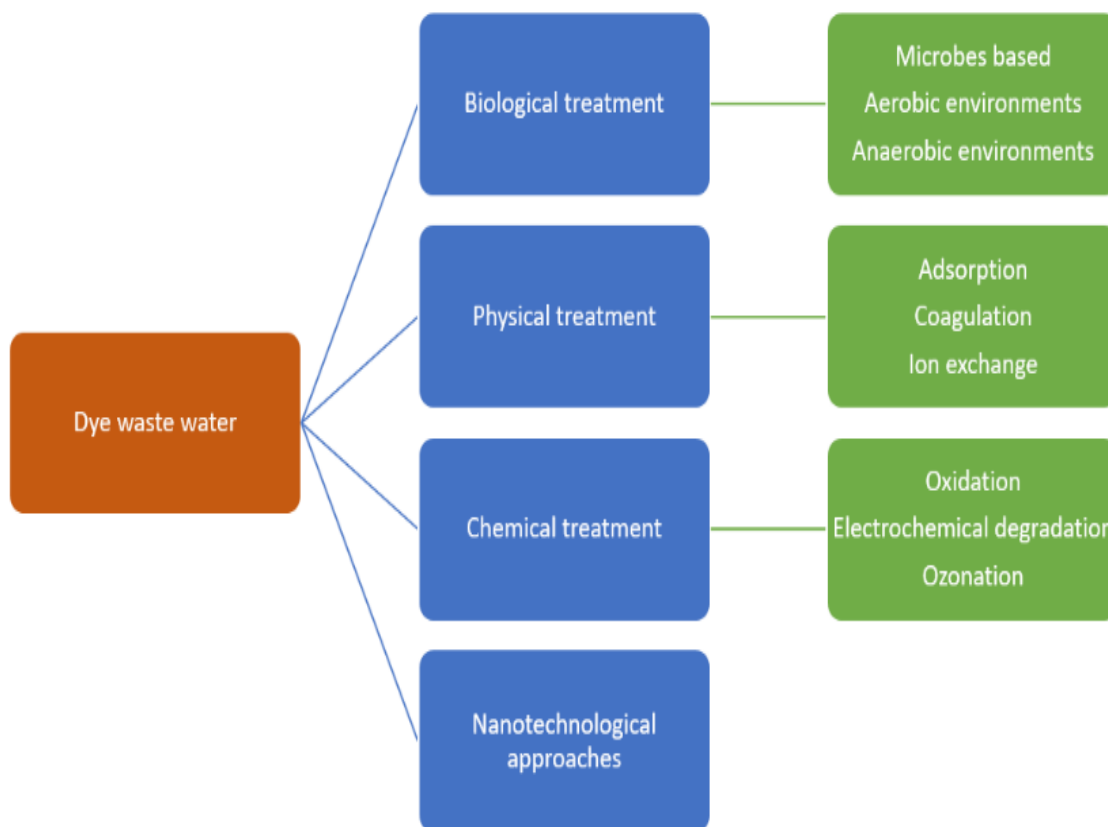


Figure 6. Various approaches for the removal of dyes in wastewater

Table 1. Classification and examples of dyes based on their structure

Classification based on structure	Examples	Characteristics
Indigoid dyes	Acid blue 71	Anionic dye, 587nm, soluble in water
Anthraquinone dyes	Reactive blue 19	Anionic dye, 594nm, soluble in water
Nitroso dyes	Fast green O	624nm, soluble in ethanol, DMF
Triarylmethane dyes	Basic violet 10B	Cationic dye, 590nm, soluble in ethanol, insoluble in xylene

Table 2. Removal of dyes using a iron oxide/CDs hybrid

Method	Catalyst dosage	Dye	Initial concentration	Performance %
Dehydration	50mg	RhB	10mg L ⁻¹	90
Dehydration	20mg	MB	20mg L ⁻¹	84
Dehydration	0.05g	MB	20mg L ⁻¹	85.6
Dehydration	0.03g	RhB	10ppm	92

5.5. Dye removal using photocatalytic degradation mechanism of CD/MO

A very effective and safe method for removing synthetic colors from wastewater is photocatalytic degradation. Among other photographic catalysts, CD/MO composites have garnered a lot of attention because of their higher light-harvesting capacity, effective carrier charge separation, and improved luminescent upon irradiation. Whenever the combined material is subjected to appropriate source of light, namely one whose light intensity equals or surpasses the bandgap energies of the metallic catalyst, two basic reactions occur simultaneously: an oxidative process supported by holes generated by light (h^+) and a reduction process accelerated by electrons generated by light (e^-) [57]. Upon irradiation, both metal oxide and CDs nanomaterials capture light. CDs may transform low-energies light or near-infrared radiation into more intense emission that can excite metal oxide semiconductors due to their adjustable bandgap and up-conversion photoluminescence characteristics [58]. Utilizing the CD@C-mTiO₂ composite, Farjadfar et al. examined this process by employing radical scavengers to determine the active species causing dye degradation. Their results revealed that the reactive species engaged in the luminescence process are hydroxyl radicals (OH \cdot), photogenerated holes (h^+) and superoxide radicals. Luminescence spectra displayed reduced recombination of electrons into holes in the composite, indicating that the inserted CDs greatly enhanced light absorption as well as significantly contributed to charge carrier separation [59]. In addition, Zhang et al. showed that CDs/N-TiO₂ photocatalysts adopt a similar mechanism. Their research highlighted how nitrogen doping reduces TiO₂'s bandgap, improving its ability to react to visible light. This alteration enhances overall photocatalytic activity by facilitating electron transport from TiO₂ to the CDs [60].

6. Applications

6.1. Removal of dyes using carbon dots with titanium oxide

A potential useful method in altering the electrical and optical characteristics of massive semiconductor components that involves doping. According to research, CDs have a narrow band gap and can absorb visible light, which makes them possible candidates for photocatalytic uses [61]. Titanium dioxide is widely recognized as a significant photocatalyst because of its exceptional efficiency under ultraviolet (UV) light. A substance must be able to modify its oxidation state without breaking down in order to be considered state without breaking down in order to be considered an effective and useful photocatalyst. TiO₂ is a preferred option for photocatalytic applications due to its remarkable physical and chemical stability, affordability in comparison to other materials for water purification, simplicity of production in laboratories, and low-toxic nature [62]. However, because of its large band gap and the quick recombination of photogenerated electron-hole (e^-/h^+) couples, which greatly reduces their effectiveness, Titanium dioxide practical use in large-scale photocatalytic processes is still restricted. To overcome these constraints and improve its photocatalytic efficacy, a number of techniques have been developed, including coupling with tiny band gap semiconductors and metal or non-metal doping [63].

Furthermore, covering TiO₂ with CDs has been considered extensively as a novel way to deal with electron-

hole pair recombination. It has been demonstrated that this technique increases the production of electron-hole pairs, facilitates electronic transitions, increases energy absorption by TiO₂, and produces reactive oxygen species when exposed to light. Consequently, the development of extremely effective photocatalysts [64]. N-doped carbon quantum dots (NCQDs) were employed by Jin et al. to embellish TiO₂ utilizing a simple hydrothermal-calcination production technique. The NCQDs improved the separation of photogenerated electron-hole pairs, increased visible light absorption, and encouraged efficient electron transfer [65]. The photodecomposition rate of methylene blue (MB) was 2.25 times faster with only 3 weight percent NCQDs (3 NCQDs/TiO₂) than with pure TiO₂. Considering a 93.1% photodecomposition rate of MB in 60 minutes and above 86% efficiency after four cycles, the 3-NCQDs/TiO₂ catalyst showed outstanding and persistent photocatalytic activity. These outcomes were attained using an initial MB concentration of 10 ppm and a catalyst dosage of 100 mg.

6.2. Removal of dyes using carbon dots with zinc oxides

Zinc oxide (ZnO) is regarded as a potential photographic catalyst, because of its exceptional catalytic performance, affordability, broad band gap (3.37 eV), and ecological compatibility [66]. However, its narrow absorption bands coupled with rapid charge carrier recombination have a major impact on its photocatalytic effectiveness. To overcome these obstacles, the development of complex ZnO nanostructures requires necessary modifications. This can be accomplished by adding carbon nanoparticles or secondary semiconductor metals [67]. The coating of ZnO nanoparticles (NPs) onto CDs provides potential to enhance overall photocatalytic performance, prevent photo-corrosion, and improve separation of charges [68]. Kumar et al. examined the creation of heterojunctions composed of GQDs painted on ZnO and their use as effective catalysts for ecological remediation. Photocatalytic ability of these heterojunctions in breaking down colored contaminant MB dye were main focus of this research [69]. Heterojunction composed of 2 weight percent GQD (ZGQD) had greatest photocatalytic capacity among investigated combinations, attaining almost 95% destruction of MB. The effective carrier charge separation that lowers rate of recombination at photocatalyst junctions is responsible for increased photocatalytic effectiveness of ZnO-GQD heterojunctions. Its large specific surface area (353.447 m²g⁻¹) and enhanced light absorption from ultraviolet to visible region.

6.3. Removal of dyes using carbon dots with iron oxides

The majority of iron oxide phases are magnetite and maghemite, which are distinguished by remarkable physicochemical qualities including huge surface area, environmental compatibility, strong magnetic behavior, low toxicity, and affordability. Among transition metal oxides, Fe₂O₃ has attracted a lot of attention because of its unique properties [70]. Furthermore, many functional groups of CDs act as efficient locations for immobilizing metal nanoparticles, improving the parent materials stability and catalytic activity [71]. Especially, CDs has shown potential in enhancing the photocatalytic activity of materials based on iron oxide [72]. Sajjadi et al. produced Fe₃O₄ nanoparticles modified with graphene quantum dots (NS-GQDs) doped with sulphur and nitrogen, and assessed their effectiveness as

a catalyst. The decolorization efficiency of the NS-GQD-decorated Fe₃O₄ (NS-GQD/Fe₃O₄) nanoparticles was much higher at 99.0% instead of 43.7% for the pure Fe₃O₄ nanoparticles. With a dopant concentration of 3 weight %, the best catalytic performance was achieved. At an initial pH of 8, the maximum sonocatalytic decolorisation efficiency of 99% was reported. The starting MB concentration in this trial was 20 mgL⁻¹, and the catalyst dosage was maintained at 1 g L⁻¹ (Table 3) [73].

7. Conclusions

The incorporation of CDs in photocatalysis has attracted a lot of interest in the scientific community since their discovery in 2004, mainly because of their ability to bond with different substantial metal oxides for efficient dye extraction. This paper reviews current developments, highlights design methodologies for CD-derived metal oxide catalysts, and examining photocatalytic activity in decomposing a variety of dyes. Additionally, it highlights effects of these pollutants on the ecosystem and human health by analyzing many dyes present in water and their related toxicology. Therefore, it is necessary to decrease or eradicate dye pollution in water supplies. By using their strong qualities, CDs mixed with metal oxides constitute hybrid materials for their removal. Performance of separate components is surpassed by synergistic benefits resulting from this collaboration. Previous research shows that efficacy of photocatalytic systems is strongly influenced by a number of technical aspects, such as modification of band structures and optical characteristics of composite materials. Despite innovative construction of CD-derived photocatalysts and their improved catalytic efficiency, a number of challenges in this field that requires more investigation. Notably, the direct use of raw CDs as catalysts is frequently researched and neglected, despite fact that CDs have a variety of functions in increasing the photocatalytic activity of metal oxide systems.

According to recent studies, small band gap of CDs between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) makes them highly promising as independent photocatalysts. The CD/metal oxide hybrid materials were employed to remove hazardous dyes from wastewater offers both considerable hurdles and interesting prospects. At extremely low concentrations, dyes are physically complex, very durable, and highly poisonous, causing major dangers to aquatic ecosystems and human health. Although CD/metal oxide hybrids have improved photocatalytic and attractive properties, their practical use is restricted by problems such as minimal durability, low sustainability, disruption from existing pollutants, and challenges in generating effective synergy between components. Furthermore, cost-effectiveness and scalability of existing synthesis techniques continued to be significant obstacles. However, promising future prospects include the advancement in green and scalable synthesis techniques, the fabrication of interface-functionalized and coated CDs to maximize transfer of charge, and the introduction of versatility (e.g., membrane integration or magnetic recovery) to boost practical applications. In order to guarantee the harmless and efficient use of these materials in water treating plants, a comprehensive environmental risk assessment and a better mechanical knowledge are also important.

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