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Functional Oxides Nanomaterials for the Removal of Dyes

Isaac John Umaru & James Eneji Agwu
Federal University Wukari, Taraba State, Nigeria
umaruisaac@gmail.com

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Abstract

The elimination of dyes from wastewater represents a critical environmental challenge, as dye effluents from industries such as textiles, printing, and paper production pose significant risks to ecosystems and human health. Conventional treatment methods are often limited in efficiency and cost-effectiveness, necessitating the exploration of advanced alternatives. This review analyzes the application of functional oxide nanoparticles in dye removal, with particular focus on metal oxides such as titanium dioxide, iron oxide, and zinc oxide, as well as mixed metal oxides. These nanomaterials exhibit exceptional adsorption and photocatalytic properties due to their large surface area, tunable surface chemistry, and distinctive surface characteristics, which enhance dye removal efficiency. The adsorption mechanisms involve electrostatic attraction, hydrogen bonding, and π–π stacking interactions, while photocatalytic degradation relies on the generation of reactive oxygen species under light irradiation, allowing complete dye mineralization and minimizing harmful intermediates. Despite these advantages, further research is needed to evaluate long-term stability, recyclability, and potential ecological impacts, alongside the development of cost-effective synthesis methods and innovative nanomaterials with improved performance.

Keywords: Functional Oxides; Nanomaterials; Wastewater Treatment; Dye Removal; Photocatalysis

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Introduction

Water is a vital resource, essential for sustaining life. Despite 71% of the Earth's surface being covered by water, which comprises around 97.5% saline water and only 2.5% freshwater, a tiny 0.007% of all water is accessible for consumption [1]. Industrial waste primarily contaminates water, containing different colours and other contaminants [2,3]. Dyes have been widely utilised in the textile, leather tanning, cosmetics, pigment, and various other sectors [4,5]. The existence of these dyes in the hydrosphere constitutes a substantial pollution source due to their visibility at minimal concentrations and their recalcitrant nature, which can pose significant threats to aquatic life by diminishing sunlight penetration and resisting photochemical reactions. Their waste products produced during dyeing operations are toxic, mutagenic, carcinogenic, and elevate chemical oxygen demand (COD) and biochemical oxygen demand (BOD) levels in aquatic environments, hence endangering human health. [6,7]. Consequently, it is imperative to remove colours from wastewater efficiently before their ultimate release into the environment. Significant volumes of dyes are released into the environment from diverse industrial processes, presenting issues for environmental scientists. Numerous technologies such as membrane separation, improved oxidation, flocculation-coagulation, electrochemical approaches and aerobic or anaerobic treatment have been used in the removal of colours from wastewater [8,9]. Therefore, there is an urgent demand to create innovative and low-cost techniques for the removal of dyes. Adsorption, recognised as a very effective technique for wastewater treatment, has been successfully utilised to diminish dangerous inorganic and organic contaminants in effluents [10]. The superiority of adsorption in contrast to other methods is its sludge-free operation and complete removal of colours even from dilute solutions [11]. A variety of adsorbents have been reported for the practicality of low-cost adsorbents to remove colours from wastewater. Some of them are chitosan, wheat straw, wood hull, banana and orange peels, oil palm ash, wasted tea leaves, degreased coffee bean, mango peels, rice husk, palm kernel shell, coconut shell and fly ash [12-23]. However, the application of these adsorbents has been restricted because of their separation difficulties. Thus, tremendous efforts are needed to exploit novel promising adsorbents. The advancement of nanotechnology has received considerable interest from many experts in the scientific domain. The increased interest in nanoscience implies that the potential of nanomaterials (NMs) is undiscovered and yet to be addressed. Nanomaterials are of significant relevance that encompass structures and devices with sizes varying from 1 to

100 nm. Because of their specific features, wide surface area and quantum size effects, these materials can provide new opportunities (such as strong adsorption, increased redox, and photocatalytic activities) to remove pollutants in wastewater [24,25]. A range of nanomaterials i.e. nano metals, nano metal oxides, graphene or graphene-based nanomaterials and polymer-based nanomaterials, have been widely employed in the removal of pollutants in wastewater [26,27]. The results demonstrated that these nanoparticles displayed a high adsorption capacity. This review contains three main elements. Firstly, the types and toxicity of dyes are described. Then, the numerous sorts of methods employed in wastewater treatment are discussed. Thirdly, the adsorption of various dyes from aqueous solutions by a wide range of nanomaterials is elaborated and their maximum adsorption capacity on various dye adsorption is compared. Finally, a summary and prognosis in the form of findings and recommendations are presented for their full-scale applications.

Dyes

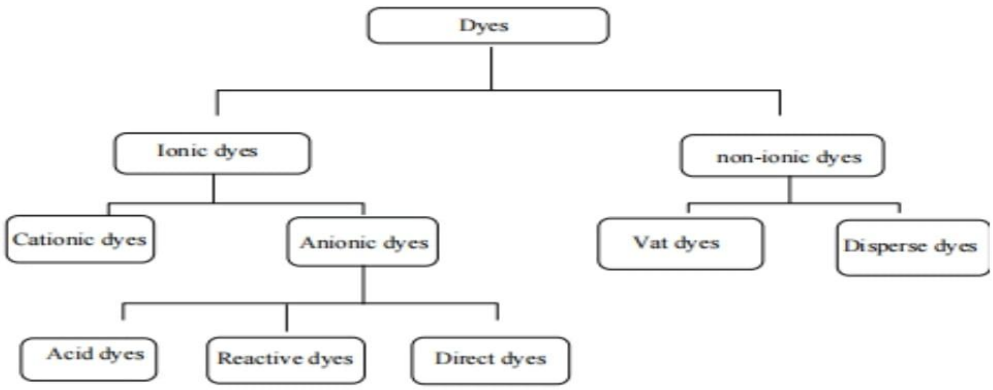


Figure 1: Classification of dyes based on ionic exchange [31]

Dyes are generally organic chemicals that can bind to the surfaces of fabrics to impart a colour [28]. Dyes can be classified according to their ingredients, colours and applications, of which application is the most generally used approach for dye categorisation [29, 30]. Azo dyes, anthraquinone dyes, phthalocyanine, indigoid dyes, nitroso dyes, nitro dyes and triarylmethane dyes based on the classification of the

chemical structure are presented in Table 1. Azo, anthraquinone and triarylmethane dyes are the most important classes among these dyes

1.1. Classification of dyes based on chemical structure

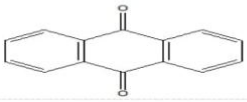
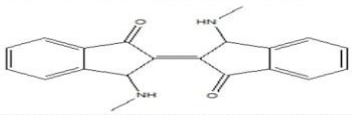
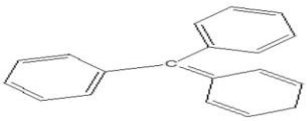
Class	Chromospheres	Examples
Azo dye		Reactive black 5
Anthraquinone dyes		Reactive blue 4
Indigoid dyes		Acid blue 71
Nitroso dyes		Acid green 1
Nitro dyes		Acid yellow 24
Triarylmethane dyes		Malachite green

Table 1: Classification of Dyes Based On Chemical Structure

Table 1 [31] shows the classification of dyes according to their many applications: vat, mordant, and dispersed dyes. Furthermore, based on how their particle charge dissolves in aqueous solutions, the dyes typically used in the textile industry are classified as cationic (all basic dyes), anionic (direct, acid, and reactive dyes), and non-ionic (disperse dyes) [32, 33]. Because of their intricate structure, dyes are useless when exposed to heat, light, bacteria, or oxidising chemicals. This makes it difficult for the dyes to degrade. Water dyes reduce water quality and prevent photosynthesis in aquatic plants and animals by altering sunlight penetration and oxygen solubility [34,35]. Furthermore, it can result in serious health problems for people, including elevated heart rate, vomiting, shock, Heinz body formation, cyanosis, jaundice, quadriplegia, and tissue necrosis [36].

1.1.1. Azo Dye

The bulk of synthetic aromatic dyes are azo dyes (monoazo, diazo, triazo, and polyazo). These dyes are made up of one, two, three, or more (N = N) groups that are attached to benzene and naphthalene rings that are regularly substituted with various functional groups, such as triazine amine, chloro, hydroxyl, methyl, nitro, and sulphonate [37]. Azo dyes are widely used in paper

printing, food colouring, cosmetics, pharmaceuticals, and textile dyeing. Because of their great stability, affordability, and range of colours, around 80% of azo dyes are utilised in the textile industry [38]. According to estimates, 10–15% of colours in effluents are released into receiving water bodies after becoming unbound with textile fibres [39]. For instance, one kilogramme of cotton required 30–60 g of dyestuff and 70–150 L of water to dye, and it has been estimated that up to 50% of used dyes were released into water sources either directly or indirectly [40,41]. Vinyl sulfone, chlorotriazine, trichloropyrimidine, and diflurochloropyrimidine are examples of reactive dyes, which are azo dyes that create covalent connections between their reactive groups [42]. Reactive dyes are superior to other dye classes in the textile industry due to their vivid colour, water-fastness, ease of application, and low energy consumption. Reactive dyes are therefore currently the most widely utilised. Reactive dyes, on the other hand, are carcinogenic and mutagenic, and even their intermediate products, mineralization, aromatic amine, and an arylamine have serious negative effects on people, including harm to the brain, liver, kidney, and central nervous system, and reproductive system [43].

[44,4

Microbial types	Dyes	Percentage removal (%)	Time
(4) fungi			
Phanerochaete chrysosporium	Reactive red 22	90-100	30 h
Trametes trogii	Anthraquinone blue	88	4 h
Trametes versicolor	Gryfalan black RL	59	400 min
Pleurotus sajor-caju	Reactive yellow 15	100	100 day
(5) yeasts			
Candida tropicalis	Reactive black 5	>99	48h
Kluyveromyces marxianus IBM3	Remazol black-B	98	24h
Rhodotorula rubra	Crystal violet	>99	4 day
(6) bacteria			
Shewanella oneidensis MR-1	Napthol green B	94	24 h
Pseudomonas sp.	Reactive blue 13;	83	72 h
Enterobacter EC3	Reactive black 5	93.6	36
Aeromonas hydrophila	Reactive red 141	80	24

Table 2: Decolorization of various azo dyes by different microbial types

1.1.2. Dyes of Anthraquinone

The textile industry has made extensive use of anthraquinone dyes due to their broad range of colour hues, fastness characteristics, ease of application, and low energy usage [54]. They are the second largest class of textile dyes after azo dyes. The strong affinity of anthraquinone dyes for silk and wool is another significant benefit of using them in dyeing procedures. However, anthraquinone dyes show a reduced rate of decolorisation because of their fused aromatic structure. Since the majority of these dyes are poisonous, mutagenic, and carcinogenic, there has been a lot of interest in getting rid of anthraquinone dyes.

1.1.3. Dyes of Triarylmethane

Triphenylmethane dyes are aromatic xenobiotics composed of one p-quinoid group (chromophore) and two benzene rings joined to a central carbon atom. -NH2, -NR2, and -OH are the most frequent auxochromes. These dyes, which include Bromophenol blue (BB), Crystal violet (CV), Malachite green (MG), and others, have vivid colours with classic red, violet, blue, and green hues [55]. In the textile industry, these dyes are frequently used to colour materials such as cotton, wool, silk, polyacrylon nitrile, and modified nylon. Petrol and the paper and leather industries are two more significant uses for triphenylmethane dyes [54]. They are thought to be appropriate for dyeing a variety of textile surfaces due to their poor light fastness, strong tinctorial strength, and reasonably cheap dyeing, particularly when washed slowly. Furthermore, triphenylmethane dyes have been widely used in medicine as medical disinfectants due to their antibacterial, antifungal, and antiprotozoal qualities. These dyes can also be used to control diabetes and disinfect wounds following surgery [56]. Because of their sensitivity to pH, certain of these dyes such as fluorescein, fuchsine, and phenolphthalein are employed as indicator dyes.

Method of removal of dyes

Biological, chemical, and physical approaches are the three primary categories into which a vast array of technologies have been created and used for dye scavenging [57]. Their practical applications are limited by their high capital costs, low efficiency, and

production of surplus sludge. A variety of colours in wastewater can be removed using some of these procedures, which are more effective and adaptable than others [58]. Adsorption is a preferable option since it is less expensive initially, creates benign byproducts, and eliminates dyes, even in diluted solutions [59].

1.2. Biological techniques

It has been demonstrated that using microbial approaches to treat industrial effluents is the most cost-effective and energy-efficient method. The treatment of industrial effluents has been investigated using microbial biosorption and biodegradation. Microorganisms such as bacteria, filamentous fungi, yeast, and algae can change dye molecules into less dangerous forms [60]. This characteristic of microorganisms is ascribed to the components of their cell walls (lipids and heteropolysaccharides), which are made up of various functional groups such as phosphate, carboxyl, hydroxyl, amino, and other charged groups. Strong attraction forces between the azo dye and the cell wall may result from them [61].

1.2.1. Fungal degradation of colours

It has been observed that fungi can degrade a variety of contaminants, including organic waste, polyaromatic hydrocarbons, dyes, and steroid chemicals [62]. Numerous intracellular and extracellular enzymes produce a metabolic product that gives rise to this feature [63]. The four primary genera of the white-rot fungus, which are the most commonly utilised fungi, are Phanerochaete, Trametes, Bjerkandera, and Pleurotus. Because of their nonspecific enzyme systems, the ligninolytic enzymes that white rot fungus produce are essential for the removal of dye [63]. Tien and Kirk were the first to introduce Phanerochaete chrysosporium, which is used to decolorise dyes [64]. According to Wu et al., after 30 hours of treatment, Phanerochaete chrysosporium may completely decolorise the Reactive Red 22 (RR 22) dye [44]. The degradation of dye in effluents was accomplished by a number of different white rot fungi, including Pleurotus sajor-caju, Trametes versicolour, and Trametes trogii [50, 65,66]. The lengthy hydraulic retention period needed for full decolourisation, however, is a barrier to using fungus to decolourise colours, and the preservation of fungi in bioreactors is another issue to take into account [65,66].

1.2.2. Yeast-degradation dyes

Since yeasts grow quickly like bacteria and can survive in harsh environments, even at low pH levels, they can be a desirable alternative for the removal of textile dyes, even if their decolourisation and degrading capabilities have only been the subject of a few research. According to a recent study by Yang Ok et al., Candida tropicalis can decolorise Reactive Black 5 (RB 5) by up to 99% after 48 hours [67]. Furthermore, it has been demonstrated that Kluyveromyces marxianus IMB3 bioaccumulates remazol black-B, with a clearance efficacy of 98% after 24 hours [748]. Rhodotorula rubra was found to be an excellent decolourizer of crystal violet by Kwasniewska et al. [49].

1.2.3. Bacterial degradation dyes

In the 1970s, research on Bacillus subtilis isolation culture was initiated [68]. Due to their ease of cultivation and quick growth, 0ria are ideal for mineralising and decolorising azo dyes. Azo dyes are biodegraded by a variety of bacterial species in facultative anaerobic, aerobic, or typical anaerobic environments. The reductive breaking of azo bonds (–N=N–) with the use of azoreductase enzymes typically initiates the bacterial degradation mechanism of azo dyes. Shewanella oneidensis MR-1 has a high capacity (94%) to decolourise Naphthol green B in anaerobic environments, according to Xiao et al. [50]. Similarly, Reactive blue 13 (RB 13) and Reactive black 5 (RB 5) were degraded by Pseudomonas sp. and Enterobacter EC3 after 70 and 36 hours, respectively (83.2% and 93.6%) [51, 52]. Additionally, several studies demonstrate that certain bacterial species may absorb dyes in aerobic environments. Reactive red 141 (RR 141) can be decoloured by Aeromonas hydrophila, and within 24 hours, the decolorisation can reach 80%.

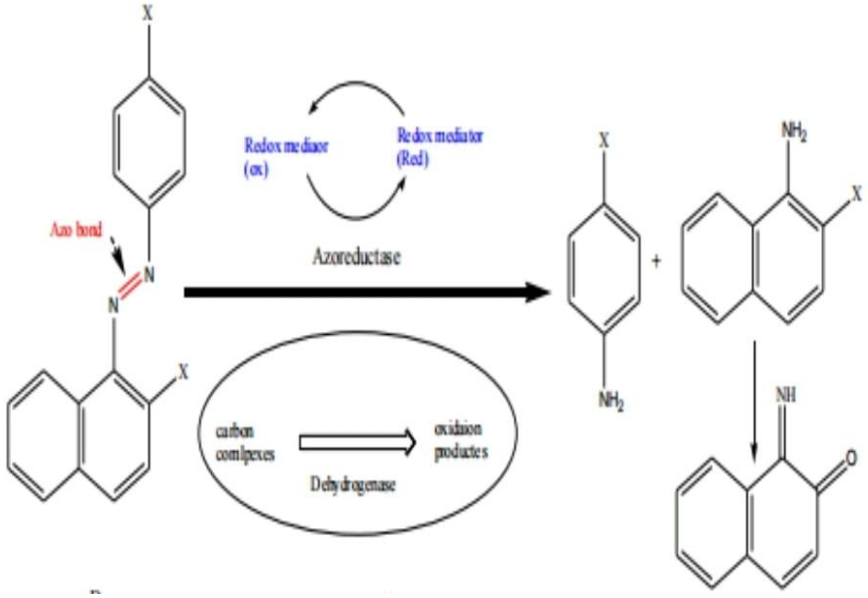


Figure 2: Mechanism of Azo dye removal by bacteria [69]

1.3. Chemical Methods

Coagulation-flocculation, ozonation, photocatalytic degradation, electrochemical therapy, and precipitation-flocculation with Fe2+/Ca(OH)2 are examples of chemical techniques. Although these chemical methods are effective at removing dyes from wastewater, overuse of chemicals can make sludge disposal challenging and increase the risk of secondary pollution. Furthermore, these methods are expensive and need a lot of electrical energy, which limits their application. Different microbial species decolourise different azo dyes.

1.3.1. Flocculation and Coagulation

As a necessary and effective method, coagulation and flocculation are typically used to purify drinking water, treat effluent from textiles, and partially lower COD. Sanchez-Martin et al. have examined this method's viability in wastewater treatment [70]. Larger particles are collected by flocculation to create microflocs, which can be eliminated in later sedimentation or flotation stages. The coagulation process destabilises colloids by adding a coagulant to neutralise the negative charges [71]. A coagulant is an agent that makes a liquid or sol coagulate. Coagulants and coagulant aids are the two primary categories of coagulants. Coagulants might be naturally occurring substances, polymers, or metallic salts. It has been documented that polyaluminum chloride and aluminium sulphate can remove dyes [72, 73]. According to Khayet et al., aluminium sulphate is more than 90% successful at removing Acid Black 210

(AB 210) when used in dosages of 40 mg/L, pH ranges of 4–8, and dye concentrations of 35 mg/L [72]. At a final pH of 10.89 and a concentration of 3000 mg/L, MgCl2 exhibits outstanding performance (98%) in the elimination of MG [74].

1.3.2. Ozonation

Ozone has a potent oxidative activity and breaks down rapidly into free radicals and oxygen. The colour is destroyed when the latter is mixed with the dyes. Both direct and indirect reactive pathways can be used by ozone to attack contaminants. The indirect ozonation is caused by highly oxidative free radicals, while the direct ozonation is associated with the ozone molecular activity. The starting dye concentration and pH have an impact on the rate at which ozone decomposes. Ozone can react with dye as an electrophile at low pH levels and can spontaneously break down into hydroxyl radicals in solution at high pH levels, which are potent, efficient, and nonselective oxidising agents [75]. According to Shaikh et al., Reactive orange 7 (RO 7), Reactive blue 19 (RB 19), and Reactive black 5 (RB 5) have nearly 100% decolorisation efficiencies [76]. For RO 7, the corresponding percentages of colour removal at pH 4, 7, 9, and 11 were 19, 50, 60, and 75%. When the pH hit 11, RB 5 began to decolorise, showing a similar pattern. The outcomes support the decolorisation process by showing that O3 breakdown and the generation of OH radicals all happened at higher pH values. According to the study conducted by Manali and colleagues, Methylene Blue (MB) degraded by up to 94.6% after 26 minutes [77].

1.3.3. Photocatalytic techniques

Due to the production of extremely reactive hydroxyl radicals, the application of advanced oxidation processes (AOPs) for the efficient oxidation of a broad range of organics and dyes has drawn particular interest in recent years. The ultrasonic process, photocatalysis, UV/H2O2, Fenton and photo-Fenton processes (Fe2+/H2O2 and Fe2+/H2O2/UV), and electrochemical processes are examples of AOPs. Because of its great efficiency and low energy usage, photocatalytic degradation is prioritised among them. There are several different kinds of photocatalysts, including titanates, perovskites, zinc oxide, and titanium dioxide. The photogeneration of charge carriers (electrons and

holes) is the reason titanium dioxide (TiO2) is utilised as a photocatalyst. Both visible and ultraviolet (UV) light can irradiate TiO2, a semiconductor with a band gap of 3.2 eV. When the photocatalyst is exposed to radiation, positive holes (h+) are created in the valance band (VB) and electrons (e-) on the surface of TiO2 are stimulated to the conduction band (CB) [78]. The electron can lower the molecular oxygen in the solution, and the positive hole can oxidise water to produce hydroxyl radicals. The following is a description of the photocatalytic mechanism: The non-toxic, low-cost, extremely stable, and reactive properties of 2TiO make it one of the most used photocatalysts. Complete mineralisation of TiO2-contaminated areas can result in non-toxic compounds like CO and HCl.

Furthermore, because of its potent oxidising ability, the reaction can occur at room temperature. To remove Direct Black 38 (DB 38), Seyyedi et al. investigated UV/TiO2 [79]. The pH of the solution has a major impact on the removal efficiency since the results indicated that the decolourisation process was pH-sensitive. Due to TiO2's 6.6 isoelectric point, the percentage of degradation is more effective at acidic pH values. The surface of TiO2 will be positively charged when the pH is below the isoelectric point, whereas the dye molecules will be negatively charged because of their sulfonic groups (SO3-). The ideal conditions for decolourising DB 38 in this process were a pH of 5, a concentration of 0.75 g/l for TiO2, a concentration of 26.56 mM for H2O2, and a dye concentration of 50 ppm calcium oxide (CaO) with varying light source radiation, with a 90% decolourisation efficiency [80]. Visible light, long UV light, and short UV light were compared. It was discovered that, in contrast to visible and long UV regions, this dye may be efficiently broken down in a brief amount of time utilising short UV light. This result showed that thanks to the advantage of higher energy, lower wavelength radiation performed exceptionally well in photodegradation. Because of its similar photodegradation mechanism to that of TiO2, zinc oxide (ZnO) has garnered a lot of attention. Because of its exceptional electrical and optical activity, excellent chemical stability, and favourable photo-electric and piezoelectric behaviours, ZnO has been used in room-temperature UV lasers, sensors, and photocatalysis [90]. To produce nano-sized particles, Zaharieva et al. synthesised ZnO using a

traditional precipitation process and then treated it with a mechanochemical technique under varying milling times [81]. Under UV light, ZnO materials were mechanochemically treated for 0, 15, 30, and 240 minutes to degrade the Reactive Black 5 (RB 5) dye. The results showed that the mechanochemically activated ZnO powder for 15 minutes had the maximum photocatalytic effectiveness (96%) while the mechanochemical treatment of ZnO materials for 4 hours had the lowest degree of degradation (82%) for RB 5 dye.

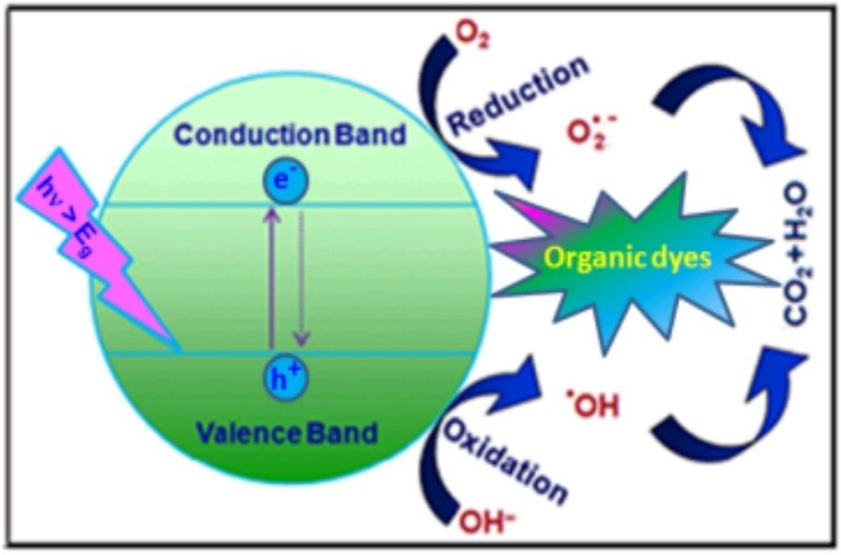


Figure 3: Schematic representation of the photocatalytic process on the surface of semiconductor nanoparticles. [82]

1.4. Electrochemical Methods

Because of its complete decolourisation, low final temperature, moderate pH range, decreased COD and BOD5, and lack of sludge formation, electrochemical techniques have garnered more attention in recent years [83,84]. While electrochemical treatment is utilised for metal recovery or compound synthesis, its applications in dye decolorisation are expanding quickly. Direct or indirect oxidation processes are used in electrochemical procedures to remove organic and hazardous contaminants. The anodic electron transfer reaction in a direct anodic oxidation method destroys the contaminants once they have been deposited on the anode surface [85]. Strong oxidants, such as hydrogen peroxide, ozone, and hypochlorite/chlorine, are produced during electrolysis in an indirect

oxidation process. The oxidant produced during the oxidation reaction process subsequently destroys the contaminants in the solution [86]. For the removal of Direct Red 80 (DR 80), Lopes et al. employed three distinct anode materials: iron, polypyrrole, and boron-doped diamond. They demonstrated a 99% removal efficiency for these materials [87].

1.5. Physical techniques

Ion exchange, adsorption, and membrane filtration (reverse osmosis, electrodialysis, and nano-filtration) are some of the physical techniques that have been developed for the removal of pollutants. The two main limitations of membrane filtering technologies are their propensity for membrane fouling and their requirement for periodic replacement. Because of their low cost, straightforward design, ease of use, high effectiveness, and lack of toxicity, adsorption techniques are a desirable substitute for treating contaminated waterways.

1.5.1. Filtration by membrane

Membrane filtration, such as reverse osmosis (RO), microfiltration (MF), ultrafiltration (UF), and nanofiltration (NF), is a sophisticated treatment technique used to eliminate colour, BOD, and COD. The basic idea behind this process is that the membrane's pore size is established by whether the particles are held on the barrier's side or pass through [88]. Particles will be prevented from passing through if they are bigger than the membrane's pore size. Because of its pore size, UF may be used to separate particles with sizes between 0.05 and 0.15 μm, which are appropriate for treating inorganic particles or organic colloids. Alventosa-de Lara et al. conducted studies to eliminate Reactive Black 5 using UF membranes [89]. The findings demonstrated that at a dye concentration of 500 mg/L, the rejection stayed above 70%. Therefore, UF membranes are not appropriate for the removal of dyes. The NF membrane has pores that are between 1 and 10 angstroms in size, which is sufficient to separate particles that are nanometres in diameter. When it comes to better flux permeation, higher retention of multivalent salts, comparatively low investment, and low operation and maintenance expenses, NF technology—a mix of UF and RO—offers advantages over UF and RO [90]. Methylene blue (MB),

Victoria blue (VB), and methyl violet (MV) colours can be effectively removed using NF in conjunction with cellulose nanocrystals [91]. Amin et al. conducted a study to examine the NF for treating Reactive Black B (RB 5) from dye-house effluents under a range of operating conditions, including temperature, flow rate, and pressure [92]. It was discovered that, with a dye rejection rate of over 99%, the ideal working conditions were 15 bar of operating pressure, 35°C, and 480 L/h of flow rate. The outcomes demonstrated that nanofiltration is a very effective method for getting rid of reactive dyes.

1.5.2. Exchange of ions

Due to their high efficiency, affordability, and appealing features, ion exchange membranes have drawn a lot of attention for use in water purification, heavy metal removal, and the treatment of textile effluents. Strong interactions between charged dyes and functional groups on ion exchange resins enable the ion exchange method to extract dyes from aqueous solutions. Anion exchange membranes, cation exchange membranes, cross-linkage membranes, and other ion exchange membranes are the four primary classifications for resins. Anion and cation membranes are the most often utilised among them for the removal of dyes. Positively charged groups like -NH3+, -NRH2+, -NR2H+, -NR3+, and -PR3+ are present in anion exchange membranes and are useful for passing anions but blocking cations. Negatively charged groups like -SO3−, -COO−, -PO32−, -PO3H-, and -C6H4O-found in cation exchange membranes permit cations to flow through while rejecting anions [93]. Muhammad et al. [94] have studied the effectiveness of an anion exchange membrane (EBTAC) in removing the anionic dye Congo red (CR) from aqueous solutions. Temperature, ionic strength, membrane dose, and contact time were among the variables whose effects were examined. The findings showed that the effectiveness of CR removal is significantly influenced by contact time and membrane dose. However, temperature and ionic strength have minimal effects on the elimination of CR. In order to remove Acid Orange 7 dye (AO7), Akazdam and Chafi expanded their research to include the use of macroporous strongly basic anion exchange resin [95]. Through a series of studies involving a number of parameters, including contact time, pH, initial dye concentration, adsorbent dosage, agitation speed, and solution

temperature, they examined the effectiveness of AO 7. The pH of the solution has a significant effect on the magnitude of this activity, according to the scientists. Up to 75% of AO7 could be nearly eliminated by the resin in 100 minutes. The findings show that, with a maximum adsorption capacity of 200 mg/g, the experimental results fit the Langmuir isotherm model quite well. Thermodynamic tests further verified that the adsorption of AO7 on the macroporous basic anion exchange resin under the studied conditions was an exothermic and spontaneous process, while adsorption kinetics demonstrated that the adsorption process followed a pseudo-second-order model.

1.5.3. Adsorption

The process by which the interface between gas and liquid molecules is focused on the surface of a porous substance is known as adsorption. Adsorbate is the substance that accumulates at the interface, while adsorbent is the substance that comes into contact with a solid [96]. Depending on how the adsorbate species is adsorbed onto the adsorbent surface, the adsorption process can be broadly classified into two types: physical sorption and chemical sorption. Through weak physical forces such as Van der Waals forces, hydrophobicity, hydrogen bonding, polarity, static interactions, dipole interactions, and π-π interactions, the adsorbate adheres to the adsorbents during physical adsorption [97]. In chemical adsorption, the force of electron exchange binds adsorbates chemically to the surface of an adsorbent. The adsorption method is a popular alternative for environmental remediation since it is more effective than other techniques for removing colours from wastewater when cost, efficiency, and design flexibility are taken into account. Adsorption also prevents secondary contamination and doesn't produce any dangerous materials. Based on the adsorption capacity, surface area, and possible cost of material reuse, an economical and environmentally friendly adsorbent was chosen for the adsorption procedure [98]. The porous structure of activated carbon (AC), which has a high surface area of 500–2000 m2/g, and its excellent adsorption capabilities for a variety of contaminants are well recognised. The source of natural resources is responsible for these exceptional qualities. However, because coal and wood are rather costly natural resources, it is essential to look for less expensive and renewable precursors. The primary

sources of AC are agricultural waste and byproducts, including chitosan, oil palm ash, banana and orange peels, wasted tea leaves, and degreased coffee beans. According to a study by Tan et al., activated carbon made from coconut husk can remove MB [99]. When the temperature was raised from 30 to 50°C, the adsorption capacity dropped from 434.78 to 384.62 mg/g, indicating that the adsorption reaction was exothermic. Equilibrium was reached at 30 hours. According to the Langmuir isotherm calculations, 434.78 mg/g is the maximum amount of MB that can be adsorbed onto an adsorbent with a comparatively high surface area of 1940 m2/g and a total pore volume of 1.143 cm3/g. Hameed et al. [100] looked into the adsorption capacity of activated carbon made from rattan sawdust. The equilibrium was reached in less than twenty-four hours, and the kinetic data were found to be consistent with pseudo-second-order kinetics. The equilibrium data demonstrated the monolayer coverage of dye molecules at the sawdust carbon's outer surface, by the Langmuir model.

1.6. Functional oxide nanomaterials for the removal of dyes

The fact that nanoscience is becoming more popular suggests that the potential of nanomaterials (NMs) is still untapped and unexplored. The elimination of dyes is a common application for inorganic nanomaterials, such as metal oxide-based or nanosized metal. High surface area and specific affinity are provided by nanosized metals or metal oxides, such as magnetic Fe3O4 [103], magnesium oxide (MgO) [103], titanium dioxide (TiO2) [104], zinc oxide (ZnO) [105], nano zerovalent iron [101], and nano zerovalent zinc [102]. These compounds, particularly metal oxides, have no secondary pollution, poor solubility, and little impact on the environment. They have also been chosen to eliminate heavy metals because of their superiority.

1.6.1. Iron nano-zerovalent

A wide range of contaminants, including nitrates, organochlorine insecticides, chlorinated chemicals, heavy metals, and dyes, can be effectively removed by nano-zerovalent iron (nZVI) particles [106–107]. In contrast to their microscale counterparts, the oxidation of the Fe core is thought to be the primary cause of the reactivity of ZVI core-shell nanoparticles. The higher

reactivity of the nanoscale particles has been ascribed to the higher density and intrinsic reactivity of their reactive surface sites. Three azo dyes, Sunset Yellow, Acid Blue A, and Methyl orange, were reported to be removed with nZVI by Rahman et al. [108]. The decolonisation of the dyes was aided by increasing the dose of nZVI particles, as seen by the removal efficiency of these three dyes in response to the nZVI dosage. However, when the dye concentration increased, the percentage of degradation dropped. The impact of the pH shift on dye removal effectiveness demonstrated that the acidic environment was conducive to adsorption. This outcome could also be explained by the iron particle electrons that are contributed to H+ ions, which results in the creation of atoms. When the pH of dyes drops, these atoms cause the conjugated system and chromophore group to cleave [109]. The outcomes showed that nZVI has a high decolorisation efficiency for Sunset yellow, Acid blue A, and Methyl orange. This substance was then employed to remove Reactive Black 5, Reactive Red 198, and Light Green, and it continues to remove these dyes with good results [101, 110]. By adding 500 mg of nZVI, the initial concentration of 100 ppm of reactive black 5 and red 198 was eliminated. Furthermore, 97% of the light green dye may be removed.

Catalyst (Nanocomposites)	Dyes	Working volume/ concentrations	Amount/ conditions	Efficiency (%), with time
$BiVO_4/CuCr_2O_4/PANI$	MB	40 mL, 10 mg L^{-1}	10 mg, 65 W, Philips lamp	95, 20 min
Ag_3PO_4/TiO_2	MB, RhB	100 mL, 20 mg L^{-1}	50 mg, 300 W Xe arc lamp	90, 42 min & 6, 42 min
Ag_2CO_3	RhB MO MB	100 mL, 10 mgL^{-1}	300W Xe arc lamp, visible light	100, 35 min 90, 15 min & 95, 30 min, respectively
$Bi_2TiO_4F_2$-12 h	RhB	60 mL, 10 mgL^{-1}	50 mg, 300 W Xe lamp	100, 150 min

e 3: The removal efficiency of Azo dyes by ZVI [89, 92, 94, 95, 99,100]

1.6.2. Nanomaterials with magnetic properties

One of the most reactive metals, iron can coordinate with other elements because it can exist in several oxidation states. Haematite (γ-Fe2O3), maghemite (α-Fe2O3), and magnetite (Fe3O4) are the three oxide types that are most frequently found in nature [111, 112]. These materials have several uses and are more re-collectable due to their high surface area-to-volume ratio, strong magnetic susceptibility, and superior biocompatibility [113]. In aqueous settings, bare magnetite nanoparticles are prone to aggregation and air-induced oxidation [114]. To eliminate Rhodamine B (Rh B), Peng et al. coated Fe3O4 nanoparticles with humic acid (HA). HA improved the stability of Fe3O4 nanoparticles and stopped them from oxidising [115]. Rh B adsorption onto Fe3O4/HA showed a maximum adsorption capacity of 161.8 mg/g and achieved equilibrium in 15 minutes, according to the data. At the ideal pH, HA-coated Fe3O4 nanoparticles were able to eliminate more than 98.5% of Rh B. Using cetyltrimethylammonium bromide as a surfactant, Chaudhary and colleagues created Fe3O4 nanoparticles by chemical precipitation to eliminate Congo red (CR), Coomassie brilliant blue R-250 (CBB), and Acridine orange (AO) [116]. With a dosage of 0.02 g and a pH of 4 for CBB and 6 for AO and CR, a good clearance efficiency was attained. Tan and colleagues investigated the adsorption capability of impregnated magnetic nanoparticles onto maize cobs for the removal of Methylene Blue (MB), and the results demonstrated favourable adsorption behaviour for MB with a removal efficiency of 99.9% [117]. Other materials including silica-based cyclodextrin (Al-CD-MNPs), polyacrylic acid, and ionic liquids can also be used to modify the surface of IONPs. These compounds were used to successfully remove Direct Blue 15, Rhodamine 6G, and Reactive Red 120 [118–119].

1.6.3. Magnesium oxide nanoparticles

Because of their high surface reactivity and adsorption capacity, nanosized alkaline earth metal oxides—in particular, magnesium oxide (nano-MgO) are an intriguing, multipurpose, and incredibly significant material that has been widely employed as a destructive adsorbent for the removal of numerous hazardous chemicals [120,121]. Because of its exceptional optical,

electrical, thermodynamic, mechanical, electronic, and unique chemical qualities, this substance is also used as a catalyst support, in toxic waste cleanup, as an additive in refractory materials, and in paint and superconducting products. Additionally, this substance was investigated as an adsorbent and bactericide [122–123]. MgO nanoparticles were demonstrated to be an efficient sorbent for Reactive Orange 122 (RO 122) and Reactive Black 5 (RB 5) in a study conducted by Jamil et al. The maximal adsorption capabilities of RO 122 and RB 5 were determined to be 333.34 mg/g and 500 mg/g, respectively [124]. Massive efforts have recently been made to create MgO with increased surface area in a variety of shapes, including tubes, wires, belts, and rods. Dhal et al. conducted research on the capacity of magnesium oxide nanomaterials in various forms, including MgO nanorods, hierarchical nanostructures, and nanoflakes, to function as adsorbents for the elimination of harmful dyes, including Congo red and malachite green [125]. The hierarchical MgO nanostructures demonstrated exceptional adsorption performance for the removal of both dyes, with maximum sorption capacities of 1205.23 mg/g and 1050.81 mg/g, respectively, which were significantly higher than those of other adsorbents, according to a comparison of three materials (MgO nanorods, hierarchical nanostructures, and nanoflakes). Based on the likelihood of its high surface area and hierarchical structures, we can explain why the hierarchical MgO nanostructures have a higher adsorption capacity than MG and CR. To improve the ability to remove dyes, nano MgO modification creates an improved composite material. Daniel and his colleagues' production of MgO-encapsulated activated carbon nanoparticles serves as one illustration. Rhodamine B (Rh B) was removed using this substance as the adsorbent [126]. With a maximum adsorption capacity of 16.2 mg/g at pH 6.75, dosage of 100 mg, and contact period of 2 hours, it was discovered that the Langmuir isotherm provides a more accurate description of the adsorption process. Moazzam et al. [127] used a modified coprecipitation approach to create rice straw charcoal/MgO nanocomposites. By altering the amount of shaking time, adsorbent dose, shaking speed, pH, temperature, and initial adsorbate concentration, several adsorption tests were conducted to examine the capacity of rice straw charcoal/MgO nanocomposite to remove Reactive Blue 221 (RB

221) from aqueous solutions [127]. It was discovered that the ideal adsorption occurred when the adsorbent dose was 250 mg, the shaking speed was 200 rpm, the pH was 7.0, the shaking period was 90 minutes, the temperature was 26°C, and the initial adsorbate concentration was 30 mg/L. This dye was adsorbed according to the Freundlich isotherm and pseudo-first-order kinetics. According to the Langmuir isotherm, the maximum adsorption capacity was determined to be 27.78 mg/g. The findings show that RB-221 can be adsorbed from aqueous solutions onto a rice straw charcoal/MgO nanocomposite.

1.6.4. Graphene oxide and graphene oxcide based

With a few layers tightly packed in six-membered rings of sp2 carbon atoms, graphene is a two-dimensional nanomaterial with exceptional properties like a high surface area (theoretically, the specific surface area is 2630 m2/g), strong mechanical, electrical, and chemical stability [128,129]. As a result, it may find use in sensors, nano-electronic devices, and nanocomposite materials [127]. Furthermore, it has a high π-π stacking interaction with the aromatic moieties found in different colours due to its graphitised basal plane structure [141]. The oxygen-rich functional groups (such as carboxyl, carbonyl, and hydroxyl groups) on the surface of graphene oxide (GO), the functionalised graphene, enable it to dissolve in water and improve its electrostatic interactions with cationic dye molecules. As a viable substrate for the creation of different graphene-based nanocomposites, GO has garnered a lot of research interest.

Adsorbents	Preparation/ modification	Adsorbate	Efficiency
Fe_3O_4 nanomaterials	NPs were synthesized by chemical precipitation method using CTAB	Acridine orange	0.056 (mol/g)
		Coomassie brilliant blue R-250	0.082
		Congo red	0.078
HA- Fe_3O_4	Fe_3O_4 NPs were modified by humic acid	Rhodamine B	Adsorption qmax-161.8 mg/g (98.5%)
Fe_3O_4 nanomaterials	Ionic liquid were used to modify Fe_3O_4 nanoparticles	Reactive red 120	166.67 mg/g
Fe_3O_4 nanomaterials	Polyacrylic-acid bound Fe_3O_4 nanoparticles, which act as core and PAA as ionic exchange	Rhodamine 6G	55.8 mg/g
Fe_3O_4 nanomaterials	Silica-based - cyclodextrin immobilized on magnetic nanoparticles	Direct blue 15	98%
MgO nano particles	Co-precipitation method	Reactive black 5	500 mg/g
MgO nano particles		Reactive orange	333.34 mg/g
MgO nanorods	Artinite and MgO nanorods prepared by precipitation method	Malachite green and Congo red	95.1% and 86.28%
MgO nanostructures	Hierarchical hydromagnesite and MgO nanostructures prepared by reflux method	Malachite green and Congo red	99.98% and 99.94%
MgO nanoflakes	Hydro magnesite and MgO nanoflakes prepared by hydrothermal method	Malachite green and Congo red	97.42%, and 92.68%
MgO nano particles	Activated carbon immobilized on MgO nanoparticles	Rhodamine B	16.2 mg/g
Rice straw charcoal/MgO nanocomposite	Rice straw charcoal immobilized on MgO	Reactive blue 221	27.78 mg/g
GO	-	Methylene blue	714 mg/g
magnetite/reduced graphene oxide	-	Rhodamine B	91%
		Malachite green	94%
nZVI/rGO	-	Rhodamine B	87.72 mg/g

Table 4: Removal of dyes by different nanomaterials [115,116,118,133,119,124,125,126,127,130,131,132]

Conclusion

The research of functional oxide nanoparticles for the removal of dyes is a promising path in resolving environmental concerns connected to water contamination. The unique qualities of these nanomaterials, such as high surface area, reactivity, and programmable surface chemistry, make them attractive candidates for efficient dye removal methods. The complete analysis of the literature shows the broad range of functional oxide nanomaterials exploited, including but not limited to titanium dioxide, zinc oxide, and iron oxide, each demonstrating significant advantages in dye adsorption and degradation. The success of these materials resides not only in their intrinsic qualities but also in the new production processes and surface changes that increase their performance. The removal processes, whether adsorption, photocatalysis or a combination of both, underline the adaptability of functional oxide nanoparticles in handling different dye contaminants.

Furthermore, the examination of various dye kinds and concentrations demonstrates the potential use of these nanoparticles across a broad spectrum of industrial effluents.

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