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# Adsorptive Remediation of Dyes Through A Novel Approach from Nanotechnology: A Comprehensive Review

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## ABSTRACT

Due to rapid industrial growth and the increased economic status of people, water sources across the globe are being significantly polluted with a wide array of effluents. Industrial, agronomic, and customary activities have led to the repeated infestation of water by discarded materials. Consequently, there is an urgent need for advanced technologies to effectively eradicate these impurities from wastewater. Among the various methods established for wastewater remediation, the adsorption process has gained remarkable significance due to its efficiency and effectiveness. The use of nano adsorbents (NADs) represents an emerging solution to these environmental issues. NADs possess exceptional physical and chemical characteristics, which enhance their applicability compared to traditional adsorbents. Their advanced grade, prominence, and excellence in various arenas make them a superior choice for wastewater treatment. Recent explorations have shown that NADs, such as carbon nanotubes, graphene, and metal and metal oxide nano adsorbents, have a pronounced and favorable impact on wastewater treatment. The focus of this review article is to provide current data and insights into the use of NADs for wastewater remediation. It aims to highlight the benefits of these novel materials and to discuss the potential areas for further improvement in this field. By exploring the latest advancements and applications of NADs, this review seeks to contribute to the ongoing efforts to address the critical issue of water pollution and to promote sustainable water management practices.

## INTRODUCTION

Rapid industrialization, together with innovation in science and technology, upgrades the standard of living, opening out towards viable economic growth and worldwide hustle (Xu et al. 2018, Nayyar 2021). The penalty of such rapid growth is an environment convoyed by considerable pollution issues (Rasheed et al. 2020). Water contamination is a severe concern that the world is facing nowadays (Afroz et al. 2014). At present, the world is facing the problem of scarcity of clean drinking water because of inadequate clean water resources (Afroz et al. 2014). Worsening of the water quality alongside the unceasing reduction in the accessibility of clean drinking water is due to the enhanced usage of water in industrial, domestic, and agronomic zones (Pimentel et al. 2004, Goel 2006). Industrial runoff encompasses many precarious and virulent pollutants severely affecting the ecosystem (McGlade et al. 2020, Sharma et al. 2021). An extensive array of waste is generated from these industries which are ultimately discharged into the water resources and, henceforth, modifying the characteristics and aesthetics of water (Gardetti & Torres 2017, Loucks & Van Beek 2017). Amongst the numerous wastes originating in these industries, colored stuff predominantly, the dyes are the supreme released contaminant (Tahir et al. 2021). To boost the aesthetics of the goods, dyes are far and wide exploited by a lot of industries such as food, textile, rubber, automobiles, cosmetics, printing and photographic sectors, pharmaceuticals, etc. (Gázquez et al. 2014, Tahir et al. 2021). As soon as the dye-laden effluents from such industries enter the water resources, it is now untimely and occasionally challenging to treat because of the complex structure and non-biodegradable character of dyes (Dhakate et al. 2020). Textile industries use dyes on a large scale. Commercially, about 0.1 million dyes are accessible globally, and around 7.0 × 105 metric tons of dyes are manufactured annually (Bharathi & Ramesh, 2013, Cockerham et al. 2022). The total dye input in textile industries is higher than 104 tons annually, and approximately 10-15% of dyes are released back into water (Husain 2006, Singh & Arora 2011). Dyes can be mutagenic, cancer-causing, and potential allergen. Therefore, the adulteration of dyes in water can be menacing for flora and fauna as well as humans (Khan et al. 2017b Ramesh & Muthuraman 2018).

## IMPACT OF DYES ON LIFE

Dyes, as pollutants, are a serious disconcertment for civilization as their intricate assemblies and non-biodegradable nature make them damaging to life (Kahlon et al. 2018). They are a precarious category of organic pollutants that are being discharged into water resources directly or indirectly (Kahlon et al. 2018). They disturb the ecosystem and are mostly carcinogenic, mutagenic, and teratogenic and also have damaging effects on the excretory, respiratory, reproductive as well as central nervous systems (Duruibe et al. 2007, Hashimi et al. 2020). They spoil the aesthetics of water bodies and curb the sunlight perforation in water, thereby affecting the aquatic flora and fauna.

Dyes are commonly classified into different categories and sub-categories (Fig. 1) on the basis of their source, well-established dye structure (nature of chromophore group), and the mode of their application (compatibility of the dye-fiber type) (Routoula 2019, Velusamy et al. 2021). Amongst the numerous classes, azo dyes are a prevalent assembly of colorants comprising almost half of all the accessible dyes used in the industries. Azo dyes are characterized by –N=N– bond where one nitrogen atom is bonded to an aromatic group (naphthalene or benzene rings) (Bafana et al. 2011, Ajmal et al. 2014, Dassanayake et al. 2021).

Furthermore, they possess amphoteric character because of the auxiliary functional groups such as carboxyl (–COOH), hydroxyl (–OH), amino (–$NH_2$), or sulfoxyl (O=S=O). These dyes may act as anionic, cationic, or non-ionic based on the pH of the surrounding medium. Classification and characteristics of azo dyes are outlined in Table 1.

Acidic dyes express damaging effects on the eyes, respiratory system, melanoma, and mutagenicity in humans (Hammam et al. 2015). Basic dyes have a noxious nature, leading to allergic complications, skin annoyance, mutations (skin carcinoma), escalation of heartbeat, moreover a rise in the prevalence of trauma, vomiting, cyanosis, icterus, tetraplegia, and tissue mortification (Afreen et al. 2018, Elgarahy et al. 2021). The amine group in these dyes is the key entity behind their toxicity (Khan et al. 2016, 2017a, 2018, Vishnu et al. 2021). Henceforth, dye remediation

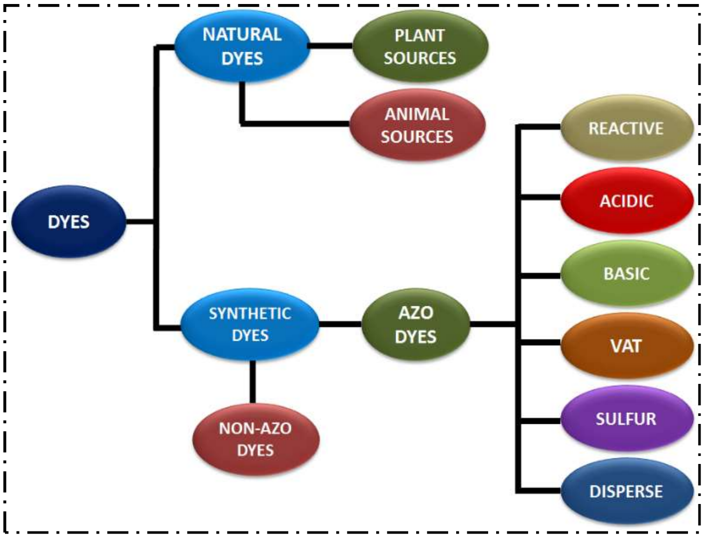

Fig. 1: Classification of dyes based on chemical composition.

Table 1: Classification of azo dyes.

| Class | Solubility Characteristic | Fiber | Fixation % | Pollutant |
|---|---|---|---|---|
| **Reactive** | • Water-soluble<br>• Anionic | • Cotton<br>• Cellulosic<br>• Wool | 60-90 | Colour, salt, alkali, unfixed dye, surfactants, defoamer, diluents |
| **Acidic** | • Water-soluble<br>• Anionic | • Cotton<br>• Nylon<br>• Wool<br>• Acrylic<br>• Protein | 80-93 | Colour; organic acids; unfixed dye |
| **Basic** | • Water-soluble<br>• cationic<br>• Applied in acidic dye baths | • Protein<br>• Cellulosic<br>• Nylon<br>• Polyester<br>• Acrylic | 97-98 | Not available |
| **Vat** | • Water-insoluble<br>• Chemically complex | • Cotton<br>• cellulosic<br>• Wool | 60-70 | Color, alkali, oxidizing, and reducing agents |
| **Sulfur** | • Water-insoluble<br>• Non-ionic | • Cotton<br>• Cellulosic | 60-70 | Color, alkali, oxidizing and reducing agents, unfixed dye |
| **Disperse** | • Water-insoluble<br>• Non-ionic | • Acrylic<br>• Modacrylic<br>• Nylon<br>• Polyester | 80-92 | Color, organic acids, carriers, leveling agents, diluents phosphates, defoamers, lubricants, dispersants |

Table 2: Pros and cons of dye-removal techniques.

| Methods | Pros | Cons |
|---|---|---|
| **Physical** (Sedimentation, filtration, floatation, coagulation, reverse osmosis, solvent extraction, adsorption) | • Good removal of a wide variety of dyes<br>• regeneration- no adsorbent loss,<br>• effective oxidation at the lab scale,<br>• economically feasible | • expensive<br>• concentrated and high sludge formation<br>• not effective for all dyes |
| **Chemical** (neutralization, reduction, oxidation, catalysis, ion exchange, electrolysis) | • simplicity of application<br>• Fenton's reagent is a suitable chemical means<br>• Ozone can be applied in its gaseous state and does not increase the volume of wastewater and sludge.<br>• No chemical consumption and no sludge buildup | • The reagent needs to be activated by some means<br>• Sludge formation<br>• Formation of by-products<br>• Relatively high flow rates cause a direct decrease in dye removal |
| **Biological** (stabilization, aerated lagoons, trickling filters, activated sludge, fungal treatment, flocculation, anaerobic digestion) | • Allows azo and other water-soluble dyes to be decolorized<br>• Certain dyes have a particular affinity for binding with microbial species<br>• Decolorized in 24 to 30 hours | • Under aerobic conditions, azo dyes are not readily metabolized<br>• Not effective for all dyes<br>• Anaerobic breakdown yields methane and hydrogen sulfide |

from wastewater before its discharge into the water sources is a serious environmental perturbation in the present hour (Madima et al. 2020, Arora et al. 2021).

## WATER MANAGEMENT TECHNOLOGIES

Physical techniques (sedimentation, filtration, floatation, coagulation, reverse osmosis, solvent extraction, and adsorption), chemical techniques (neutralization, reduction, oxidation, catalysis, ion exchange, and electrolysis), and biological techniques (stabilization, aerated lagoons, trickling filters, activated sludge, fungal treatment, flocculation and anaerobic digestion) are employed for the confiscation of dyes from wastewater (Gunatilake 2015, Carolin et al. 2017, Wu et al. 2017, Yadav et al. 2021) Such techniques are steadfast and display specific outcomes. However, on the other hand, they also have some limits like lesser efficacy, greater investment, generation of too much sludge, and high maintenance charges, making them unbefitting for economical practice.

All of these methods have their pros and cons, such as high operational /energy expenses, generation of huge expanses of sludge, and production of detrimental byproducts (Table 2).

Still, from the above-mentioned techniques for wastewater remediation, the adsorption route is the most tempting method due to its easy functioning, high efficacy, easiness, cost-efficiency, and persistence (Wu et al. 2017). Moreover, in the majority of explorations, the process is reversible, and henceforth, the adsorbents can be without difficulty recycled recurrently, making the overall adsorption process more cost-effective. Additionally, the accessibility of an immense choice of adsorbents to befit precise requisite makes it more adaptable (Creamer & Gao 2016, Wu et al. 2017). Owing to the diverse benefits aforementioned, the adsorption method has acknowledged consideration all around the environment adoring society (Liu et al. 2016).

Adsorption is the adhering of a particle superficially to a surface. The particle might be a gas, liquid, or solid (atom, molecule, or ion), and the surface might be a liquid or solid. The binding between the particle and the surface is either chemical or physical (Liu et al. 2016, Ijaz & Zafar 2021). The substances offering the surface are the adsorbents, while the entity that is attached to the surface is the adsorbate (Li et al. 2009, Farrukh et al. 2013, Dutta et al. 2019).

Adsorbate particles are associated with adsorbent via two kinds of forces, viz. physical and chemical, in the processes named physisorption and chemisorption. Physisorption is caused by feeble attractive forces amid adsorbate-adsorbent molecules, while chemisorption occurs through the formation of a strong chemical bond (Jelmy et al. 2021). An adsorbent is considered effective; constraints like surface area, porous nature, adsorption capacity, and mechanical strength ought to be extremely high alongside the viability of supplementary features like cost-efficiency, untroubled renewal, persistence, and selectiveness (Jelmy et al. 2021). A diagrammatic illustration of the elimination of dyes from wastewater using adsorption is displayed in Fig. 2. Several factors, namely, adsorbate/adsorbent interaction, surface area (adsorbent), adsorbent/adsorbate proportion, particle size (adsorbent), temperature, pH, contact time, etc., are calculated as they are accountable for the elimination rates of dyes (Gupta 2009, Banerjee et al. 2015, Mashkoor & Nasar 2020). Detailed investigations of such optimized factors are considered to be useful in the remediation of dyes efficiently and for the progress of commercial-scale wastewater treatment processes (Karimifard & Moghaddam 2018, Islam et al. 2019). Amongst the diversity of adsorbents available, activated carbon is the utmost chosen adsorbent for the confiscation of dyes owing to its outstanding adsorption capability (Ahmad et al. 2021). However, extensive application of activated carbon is constrained as it is costly (Whitacre et al. 2012, Ahmad et al. 2021). A variety of non-conventional economical adsorbent materials have been investigated by various researchers for the remediation of dyes (Crini 2006, Rafatullah et al. 2010, Dawood & Sen 2014). These encompass agricultural wastes industrial waste products, clay materials, zeolites, siliceous materials, biosorbents, biomass and others (cyclodextrin, starch, cotton), Artocarpus heterophyllus peel, Allium sativum peel, hazelnut shell, pineapple stem, longan shell, consumed tea leaves zeolite, corncobs and so on (Bouzaida & Rammah 2002, Crini & Morcellet 2002, O'mahony et al. 2002, Özacar & Şengil 2002, Crini 2003, Delval et al. 2003, Walker et al. 2003, Allen et al. 2004, Wong et al. 2004, Aksu & Tezer 2005, Hameed 2009). Consequently, the considerations have been encouraged to discover more economical and proficient replacements of activated carbon. Innate components, agronomic and industrialized trashes and bio-sorbents can be the most credible replacements (Whitacre et al. 2012).

Also, the non-conventional adsorbents displayed diversified conclusions, with several cases exhibiting high removal efficiencies as compared to the activated carbons (Gupta et al. 2013, Vieira et al. 2020).

But, the investigations for innovative and futuristic adsorbents with high dye removal efficiencies with excellent regenerability are a crucial part of current research in the water treatment field. Currently, nanoparticles (NPs) are

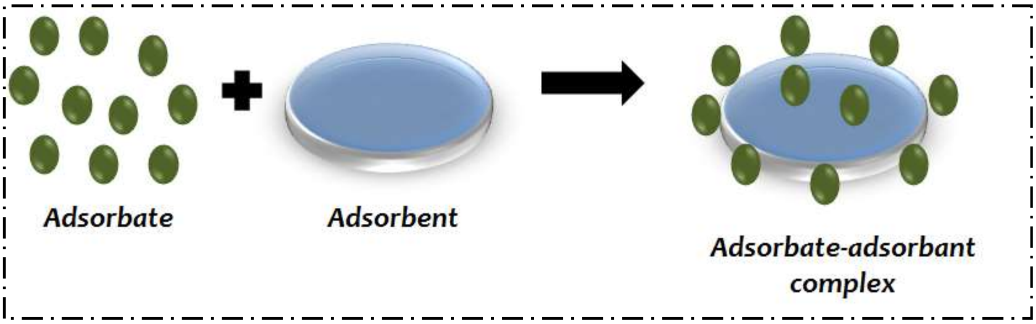

Fig. 2: Schematic representation of the Adsorption process.

being exploited for the decontamination of wastewater. These anticipated NPs are being made with distinctive assets like enormous surface area/volume ratio and surface characteristics to tackle noxious contaminants.

## IMPLEMENTATION OF NANOTECHNOLOGY IN DYES REMOVAL

Nanotechnology presents a new arena of science offering a significant part in wastewater treatment (Nasrollahzadeh et al. 201, Thangadurai et al. 2020, Shakoor et al. 2023). The word 'Nano' is extracted from the Greek term 'Nanos', meaning dwarf or exceptionally tiny. Nanoparticle size covers the 1-100 nm range and is distributed in all media (gaseous, liquid, solid). Such characteristics make them the most desirable materials for an extensive array of uses (Gangadhar et al. 2012). NPs may be categorized as inorganic, polymeric, solid lipids, liposomes, nano-crystal, nanotubes, and dendrimers (Biswas et al. 2014, Ansari et al. 2020). NPs are synthesized via physical, chemical, or biological techniques (Jameel et al. 2020). The chemical technique is significant since it requires little duration for the preparation of a variety of NPs (Ju-Nam & Lead 2008). Polymeric NPs are synthesized via the polymerization of several monomer units like methacrylic acid, acrylic esters, methacrylic, and so on (Martin et al. 2021). Synthesis of inorganic NPs might be done in the manifestation of polymers like polylactic acid and polyglycolic acid etc. (Xue et al. 2019). Presently, chemical techniques, viz., dispersion of pre-formed polymers, polymerization of monomeric units, and coacervation of hydrophilic polymers, are practically aiming for the synthesis of diverse polymeric NPs (Xue et al. 2019). Metal/metal oxide NPs might be synthesized via different physical methods, namely, evaporation-condensation, sol-gel technique, solvothermal method, chemical reduction, laser ablation, etc. Amongst several methodologies, the biological one is exceedingly effective owing to its environmentally friendly approach. This category of NPs includes those synthesized using plants, fungi, yeast, bacteria, viruses, proteins, enzymes etc., (Saratale et al. 2018, Salem & Fouda 2021).

Numerous nanomaterials have been synthesized and used for the sequestration of impurities existing in wastewater (Ata et al. 2019, Mustapha et al. 2020). They have remarkably high surface area-volume ratio, micro- or mesoporous structure, high adsorption capability, cost-efficiency, and regenerability (Chang et al. 2019). These nano-adsorbents (NADs) perforate deeply, act quickly, and possess outstanding pollutant-binding capability (Chang et al. 2019). NADs might be shaped into nanowires, nanotubes, nanofilms, and nanoparticles. In wastewater management, several practically viable and effective NADs are recognized as having characteristic features for successful decontamination of wastewater (Sharma et al. 2009, Harja & Ciobanu 2020, Janani et al. 2022). Outstanding removal efficiencies of NADs have been published in the literature regarding the confiscation of dyes from wastewater (Ahmed et al. 2020, Essekri et al. 2021, Ansari et al. 2023, Afridi et al. 2023). NADs can be classified into different categories based on their application in adsorption techniques, namely, magnetic, nanostructured mixed oxides, and metallic/metallic oxides (Ahmadi et al. 2017). In addition to these, nanotubes, nanosheets, and carbon/silicon/polymer nanoparticles are a few NADs utilized for the adsorptive removal of dyes from wastewater (Ahmadi et al. 2017).

Wastewater treatment methods using nanotechnology are hopeful approaches to overcome the key hurdles in water treatment technologies.

## NANO-ENGINEERED ADSORBENT

Prerequisites for an efficient adsorbent are the constraints like higher surface area, highly favorable porosity, greater absorptivity, and eminent mechanical strength, as well as supplementary features like cost-efficiency, easy regenerability, viability, and selectivity (Mahfoudhi & Boufi 2017). Numerous adsorbents are being used for wastewater treatment, which mainly include those obtained from agronomic, domestic, and industrialized wastes, polymeric materials, and organic as well as inorganic substances (Mahfoudhi & Boufi 2017). However, the adsorbents obtained from such sources have low absorptivity (Mahfoudhi & Boufi 2017). Hence, it has become essential to discover more innovative, operative, and excellent adsorbents for wastewater treatment (Tara et al. 2020). The cost and physiognomies like particle size, homogeneous size organization, shape, crystal framework, composition, purity, stabilization, and reproducibility make the NPs appropriate for utilization in various fields like sensors, biomedical applications, and, in particular, in water treatment. The utilization of NADs in wastewater treatment opens up a new arena for the application of nanotechnology (Tara et al. 2020).

The most extensively studied NADs for wastewater treatment are carbon nanotubes (CNTs), graphene and metal oxides such as $Fe_3O_4$, $MnO_2$, $CO_3O_4$, $TiO_2$, MgO, ZnO, etc (Luo et al. 2010, Apul et al. 2013, Abas et al. 2014, Khan et al. 2016, 2015, Zare et al. 2015a, Mohammad et al. 2019, Mohammad et al. 2021a,b). They might be prepared in diverse morphological arrangements. Hereby, this review aims to update on the recent advancements in dye remediation from wastewater using NPs as efficient adsorbents along with the viewpoints in this part of the exploration.

## CARBON-BASED NANOMATERIALS

Recently, NPs based on carbon, namely, graphene, fullerenes, and carbon nanotubes, have attracted noteworthy attention owing to their superlative assets, predominantly vast surface area, porosity, thermal and mechanical strengths, and removal efficacy (Bhatnagar & Minocha 2006 Mauter & Elimelech 2008, Shakoor & Nasar 2017, 2018a, Varghese et al. 2019, Shahbazi et al. 2020).

### Carbon Nanotubes (CNTs)

Sumio Iijima, a Japanese physicist, discovered CNTs in the arc evaporation method (Ajayan et al. 1993). Apart from the arc evaporation method, several other methods, namely, laser ablation, flame synthesis, chemical vapor deposition, and electrolysis, have been described for the synthesis of CNTs (Terrones 2003, Farhat & Scott 2006, Nayeri & Jafari 2024). It is worth mentioning that arc evaporation, chemical vapor deposition, and laser ablation procedures are widely used for the synthesis of CNTs (Farhat & Scott 2006). The CNTs comprise graphene/graphite sheets that wind up in a tube shape having a diameter in the nanometer range and length in the micrometer range. The ends of CNTs are capped with a hemisphere of the fullerene-like entity. The hollow-tiered structures of CNTs offer a high surface area as well as high porosity. This unique assembly of CNTs possesses mechanical strength, electronic as well as thermal stability. They might be metallic or semiconducting depending upon the class of chiral characteristics (chiral angle between carbon hexagons and tube axis) (Aqel et al. 2012, Gusain et al. 2020).

CNTs have been substantially applied for the remediation of dyes in wastewater treatment as a result of their high absorptivity for dyes(Gusain et al. 2020, Thakur et al. 2024).

CNTs are classified into single-walled CNTs and multi-walled CNTs (Fig. 3). Single-walled CNTs are comprised of single graphene sheets that wind up in a cylinder whereas multi-walled CNTs are made of coaxial piling of graphene sheets to form of a cylinder with the adjoining sheets being adhered by weak van der Waals forces with an interspacing of approximately 0.34 nanometers (Aqel et al. 2012).

Multi-walled CNTs have been exploited for the adsorptive elimination of single dye systems such as Congo red (Zare et al. 2015b), methyl orange (Yao et al. 2011), blue 116 (Vuono et al. 2017), red 159 (Vuono et al. 2017) and yellow 81 (Vuono et al. 2017), maxilon blue (Alkaim et al. 2015), reactive blue 4 (Machado et al. 2012), reactive red M-2BE (Machado et al. 2011), direct blue 53 (Prola et al. 2013), ponceau 4R (Ferreira et al. 2017), allura red (Ferreira et al. 2017), etc. The adsorption behavior of multi-walled CNTs on cationic-anionic binary dye systems was also studied (Ma et al. 2018). Multi-walled CNTs were utilized for adsorptive remediation studies of acid red 183 and methylene blue in an aqueous solution. It was observed that multi-walled CNTs possess higher adsorptive attraction towards methylene blue as compared to acid red 183 in single as well as dual-dye systems (Ma et al. 2018).

The maximum adsorption capacity in the case of mono-dye set-up was obtained to be 59.7 for methylene blue (cationic) and 45.2 mg.g$^{-1}$ for acid red 183 (anionic).

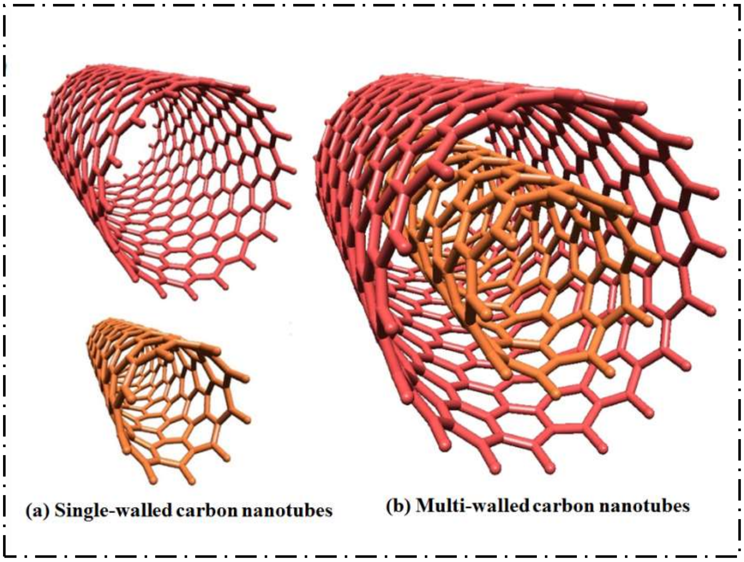

Fig. 3: Schematic representation of single-walled and multi-walled CNTs.

For dual-dye set-up, keeping the concentration of acid red 183 constant (20 mg/L), an increase in adsorptive removal of both the dyes was observed with the increase in methylene blue concentration, suggesting a synergistic effect. On the other hand, by maintaining a constant methylene blue concentration (10 mg/L), adsorptive removal of methylene blue was found to be reduced with increasing acid red 183 concentrations, while acid red 183 exhibits a rising inclination in adsorption capacity (Wang et al. 2012, Shakoor & Nasar 2016).

Functional modifications of CNTs have been commenced for the introduction of different functional groups, thereby providing fresh adsorption sites (Wang et al. 2012). Many research explorations have confirmed the enhanced elimination of dyes from wastewater by modified CNTs using various functional groups. Such functional modifications lead to the reduction in the accumulation of CNTs, escalation of adsorption, durability, selectiveness, and affinity for pollutants in wastewater (Gupta et al. 2013, Gupta & Saleh, 2013). Amongst the various functionalization methods, oxidation is the simplest mode for the introduction of –OH and C=O groups to the walls of CNTs. Multi-walled CNTs obtained after oxidation were more efficient for the elimination of methyl red (MR) and bromothymol blue (BB) in water (Ghaedi & Kokhdan 2012, Sadegh et al. 2017). An adsorption capacity of 41.63 mg.g$^{-1}$ was reported by Yao et al. for the elimination of MB on CNTs at 333 K (Yao et al. 2010). Similar experiments were executed by Shahryari et al. (Shahryari et al. 2010) on multi-walled CNTs with high surface area (280 m$^2$ g$^{-1}$) in comparison to the CNTs (160 m$^2$ g$^{-1}$) investigated by Yao et al. An adsorption capacity of 132.6 mg.g$^{-1}$ at 310 K was observed for MB dye. The adsorption capacities are also influenced by the experimental constraints and kinds of adsorbents. Relative adsorption capacities of orange II dye from aqueous solution by the utilization of multi-walled CNTs and carbon nanofibers adsorbents were calculated in batch experimentations by Rodríguez et al. (Rodríguez et al. 2010). It was observed that the adsorptive elimination of anionic orange II on multi-walled CNT (77.83 mg.g$^{-1}$) was a little higher than carbon nanofiber (66.12 mg.g$^{-1}$).

CNT-impregnated chitosan hydrogel (CS/CNT) beads were synthesized to investigate the elimination of Congo red dye. CS/CNT beads revealed a higher maximum adsorption capacity (450.4 mg.g$^{-1}$) as compared to the chitosan without impregnation (200 mg.g$^{-1}$, Langmuir isotherm model) (Chatterjee et al. 2010). A unique type of CS/CNT beads was synthesized by (Chatterjee et al. 2011) by treating multi-walled CNTs with sodium dodecyl sulfate to upgrade mechanical characteristics (Chatterjee et al. 2011). These unique CS/CNT beads revealed a maximum adsorption capacity of 375.94 mg.g$^{-1}$ for Congo red dye (Chatterjee et al. 2011). It is worthy of mentioning that CNTs might be consequently generated on a macroscale by different methodologies to reduce the production cost thereby increasing their subsequent consumption in environmental safeguard administration. Utilization of CNTs in wastewater treatment is anticipated to be a revolution in upcoming research (Hussain et al. 2024).

## Graphene

Graphene comprises one or more layers of carbon atoms adhered by weak Van der Waal forces and p–p stacking associations with a distinctive 2D assembly and tremendous mechanical, thermal, and electrical characteristics (Shahryari-ghoshekandi & Sadegh 2014, Rajabi et al. 2019). Several studies were conducted employing graphene and graphene oxide for the elimination of dyes from wastewater. For modification of the physico-chemical properties, reduced graphene oxide (rGONSs) and graphene oxide nanosheets (GONSs) were prepared by incorporating them in composite fragments. rGONSs, as well as GONSs layers, have large aspect ratios with huge electronic surfaces providing powerful intermolecular forces amongst the adsorbate molecules (Denis & Iribarne 2012). Owing to the exposed layered arrangement, rGONSs show significantly accelerated adsorption kinetics as compared to the CNTs (Ji et al. 2013, Yu et al. 2014).

Amongst the various carbon-based nanomaterials (Activated carbons, single-walled CNTs, and multi-walled CNTs), rGONSs showed enhanced absorptivity of two synthetic organic compounds (phenanthrene and biphenyl) in an aqueous medium (Thakur & Kandasubramanian 2019). Also, rGONSs are comparatively low-priced compared to single-walled CNTs. rGONSs were employed for the elimination of cationic red X-GRL (Li et al. 2011c), methylene blue (Li et al. 2011°, Yang et al. 2011), methyl orange (Li et al. 2011b), Congo red (Li et al. 2011b) from aqueous solutions. The maximum adsorption capacities for p-toluenesulfonic acid, 1-naphthalenesulfonic acid, and methylene blue on GNS goes to 1430, 1460, and 1520 mg.g-1 at 303 K, respectively, highest amongst all nanomaterials calculated so far (Wu et al. 2011). The abundance of functional groups with oxygen superficially on graphene oxide nanosheets was described as executing an essential part of the adsorption process. Relative to activated carbons and CNTs, graphene oxides and graphene nanosheets exhibit a stronger adsorption affinity for dyes in wastewater.

## Metal Oxide-Based Nanomaterials

Nanomaterials based on metal/metal oxides are a class of inorganic nanomaterials extensively utilized to confiscate dyes from wastewater. Metal oxides have negligible environmental effects, little solubility, and do not contribute to secondary pollution. Nowadays, zero-valent iron, iron oxides/hydroxides ($Fe_3O_4$), titanium oxide ($TiO_2$), zinc oxide (ZnO), and copper oxide (CuO) nanoparticles as well as in its composites form have been employed as adsorbent for the confiscation of dyes from wastewater (Khan et al. 2019b, 2019a). In recent times, zero-valent iron was synthesized by Rahman et al. (Rahman et al. 2014) with a borohydride chemical reduction method and is being employed for the elimination of azo dyes from wastewater. (Arabi et al. 2013) carried out kinetic and thermodynamic investigations for eliminating vat green dye from wastewater using zero-valent iron NADs. Diverse methodologies, namely, oxidation, reduction, disproportionation, hydrolysis, sol-gel, high-pressure hydrothermal, and co-precipitation, have been employed to synthesize iron oxide nanoparticles (Lu et al. 2006, Teja & Koh, 2009, Layek et al. 2010). Iron oxide and zero-valent iron composite NADS have also been synthesized and exploited for the elimination of precarious toxic pollutants from wastewater (catalytic and magnetic character of such NADs exhibited greater efficiency for dye confiscation). Iron being the most prevalent element in the earth crust and triviality of its resources and ease of production makes ferric oxides an economical adsorbent for dye adsorption.

Novel magnetic $Fe_3O_4$@CNADs were synthesized and employed for the confiscation of methylene blue and Congo red (Zhang & Kong 2011). The maximum adsorption capacity was found to be 44.38 and 11.22 $mg.g^{-1}$ for methylene blue and Congo red, respectively. Improvements in the iron oxides and zero-valent iron nanoparticles were also made by mingling such particles with different organic and/or inorganic constituents.

An adsorbent with a size in the nano range having an enormous surface area might proficiently confiscate dyes. However, the consumed nanoparticles are hard to isolate afterward, and continuing exposure might cause toxicity altogether. By utilizing magnetic iron oxide nanoparticles, such difficulties can be overcome. Nevertheless, additional efforts are desired to make the procedure extra efficient and innovative. The maximum adsorption capacity of magnetic zero-valent iron nanoparticles prepared by the coprecipitation method and loading on Arabic gum was found to be 14 $mg.g^{-1}$ for methylene blue dye (Alzahrani 2014). A novel bi-metallic Fe-Zn nanoparticle prepared by co-precipitation technique has been employed for the confiscation of malachite green and Congo red from wastewater by Gautam et al. (Gautam et al. 2015).The maximum adsorption capacity of the adsorbent was found to be 21.74 $mg.g^{-1}$ for malachite green and 28.56 $mg.g^{-1}$ for Congo red. The polypyrrole-coated magnetic $Fe_3O_4$ nanoparticle (PPy@$Fe_3O_4$) was also utilized as an adsorbent to eliminate synthetic textile dye RB19. The maximum adsorption capacity of PPy@$Fe_3O_4$ for RB19 was observed to be 112.4 $mg.g^{-1}$ (Shanehsaz et al., 2015). L-arginine-functionalized $Fe_3O_4$ magnetic nanoparticles ($Fe_3O_4$@L-arginine) were synthesized, and the removal efficiency for Reactive Blue 19 was observed to be 96.3% under optimum conditions. The adsorption mechanism obeyed pseudo-second-order kinetics and Freundlich isotherm (Dalvand et al. 2016).

Apart from iron, other metal-based nanoparticles and their composites have also been synthesized and exploited for dye remediation, showing tremendous removal efficiency for various types of dyes. Cupric oxide (CuO) nanoparticles synthesized by numerous procedures were employed for the removal of various dyes such as MO, MB, crystal violet (CV), CR, trypan blue (TB), etc. (Mustafa et al. 2013, Mekewi et al. 2016, Sasikala et al. 2016, Taufik & Saleh 2017, Shakoor & Nasar 2018b, 2019).

Mustafa et al. (Mustafa et al. 2013) synthesized the cupric oxide (CuO) nanoparticles using the precipitation method and employed it for the adsorptive studies on MB dye, accomplishing a removal efficiency of 88.9%. Similarly, Mekewi et al.(Mekewi et al. 2016) synthesized CuO nanoparticles and chemically activated them with montmorillonite clay, and used them for the removal of MB dye from an aqueous solution.

Sol-gel method was adopted to synthesize the iron (II, III) oxide/zinc (II) oxide/copper (II) oxide ($Fe_3O_4$/ZnO/CuO) nano-composites. The $Fe_3O_4$/ZnO/CuO nanocomposites with different amounts of CuO nanoparticles were used for photocatalytic degradation of MB dye under UV/vis light and ultrasound arrangement. Faster degradation of MB was observed using a higher quantity of CuO in visible light and also with a lower quantity of CuO in UV light (Taufik & Saleh 2017). Adsorptive studies were carried out on cerium-loaded CuO NPs by Sasikala et al. (Sasikala et al. 2016) for the remediation of azo dyes such as MO and TB in UV radiation. Silver nanoparticles-loaded AC was used as an adsorbent by Karimi et al. (Ghaedi et al. 2013) for the elimination of MO dye. In their study, it was observed that an increase in the pH of the dye solution was accompanied by enhanced adsorption of MO. The removal efficiency increased from 67.3 to 98.8%, with a rise in pH from 2 to 5. Mady et al. (Mady et al. 2017) prepared Ag-$ZnFe_2O_4$@ reduced graphene oxide (rGO) nanocomposites by a one-pot microwave-assisted self-assembly technique. The Ag-$ZnFe_2O_4$@rGO nanocomposites thus obtained with a 15.2 weight percentage of rGO exhibited brilliant adsorption characteristics and high photocatalytic activity for the elimination of degradation of rhodamine B (RhB), MB, and MO dyes. The Ag-$ZnFe_2O_4$@rGO nanocomposite can be recovered straightforwardly with a simple magnet and can be used five times with no substantial reduction in its photocatalytic activity. The Ag-$ZnFe_2O_4$@rGO nanocomposite catalyst can also be employed for the confiscation of hard-to-degrade unwanted constituents because of its high proficiency in both UV and visible light and its excellent regenerability.

Yang et al. (2016)synthesized novel silver-containing vanadate semiconductor nanorod ($Ag_2ZnV_4O_{12}$ nanorod) was prepared using the sol-gel process, and the photocatalysis was explored by photodegradation of RhB dye excited by the light wavelength higher than 420 nm.

Ni-doped ZnO nanoparticles were synthesized by a simple low-precipitation method at low temperatures and were exploited for the degradation of anionic Fast Green (FG) and cationic Victoria Blue (VB) dyes (Saharan et al. 2015), synergistic influence of Ni-doped ZnO nanoparticles and ultrasonication leads to practically complete mineralization of both FG and VB dyes in merely 5 minutes of contact time in the manifestation of light. The recovered nanoparticles were utilized yet again to degrade the same dyes recurrently under ultrasonic irradiation. The sonodegradation efficiency for FG and VB was found to be 96% and 94%, respectively, in the first 2 cycles. After that, only a slight decrease in the catalytic efficiency was observed. Cobalt ferrite (CoFe2O4) nanoparticles prepared and functionalized with an amine (-NH2) group were employed for the adsorption studies on Direct Green6 (DG6), Direct Red 80 (DR80), and Acid Blue 92 (AB92) dyes. The amine group was introduced to enhance the adsorption activity of the CoFe2O4 nanoparticles. The adsorption was quite fast, and adsorption equilibrium was attained in about 15 minutes. Experimental outcomes indicate that the system obeys the Langmuir adsorption isotherm model equation and fits better than the other equations (Yavari et al. 2016).

A thin-film $TiO_2$-coated nano-structured template was synthesized by metal-assisted wet etching of Si. This was used as a substrate for the deposition of a 10 nm thick film of $TiO_2$ by atomic layer deposition.is studied by dye degradation in water. The photocatalytic efficacy of this nanostructured template was evaluated by the degradation of two dyes in aqueous solution, namely, MB and MO (Scuderi et al. 2014).

The nanostructured $TiO_2$ revealed that the photo-degradation reaction rate is approximately 3 (for MB) and 12 times (for MO) as compared to the rate of $TiO_2$ flat film (Scuderi et al. 2014). New photoactive composites, Cu2O/TiO2 nanoparticles in novel inorganic geopolymer matrix altered by cetyltrimethylammonium bromide (CTAB), were synthesized to efficiently confiscate MB dye from aqueous solution. The mechanism of the removal of dye involves a combination of adsorption (under dark conditions) and photodegradation (under UV radiation). MB adsorption in the dark obeys pseudo-second-order kinetics and is best described by Freundlich-Langmuir isotherms. The adsorptive behavior of the CTAB-modified geopolymer-centered composites is far superior to the ones based on unmodified geopolymer hosts. The most effective composite is the one containing 5-weight percent Cu2O/TiO2 in a CTAB-modified geopolymer host. These composites set up a new class of materials with outstanding potential in environmental protection applications (Falah et al. 2016).

Glutaraldehyde cross-linked magnetic chitosan nanoparticles (GMCNs) not only exhibited outstanding adsorptive behavior for food dyes but also exhibited small cytotoxicity. Adsorption features of FD&C Blue 1 and D & C Yellow 5 in aqueous solutions by GMCNs were carried out. The adsorption mechanism was better depicted by the pseudo-second-order kinetics and the Langmuir adsorption isotherm model. Maximum adsorption capacities of GMCNs at pH 3.0 and 298K were found to be 475.6 and 292.1 $mg.g^{-1}$ for FD&C Blue 1 and D&C Yellow 5, respectively. Thermodynamic studies demonstrated that the reactions were spontaneous and exothermic (Zhou et al. 2014).

## DISADVANTAGE OF NANO ADSORBENT MATERIALS

NADs play a vital part in elucidating ecological concerns like the decontamination of wastewater due to their incredible physiochemical properties. At the same time, certain shortcomings can be acknowledged while utilizing NADs in wastewater treatment (Yaqoob et al. 2020). One key shortcoming is the probable ecotoxicity of the remaining nanomaterial in water, which might cause secondary toxic impacts and possibly hurt humans, animals, and other life forms (Wang et al. 2019, Zhu et al. 2019, Sardar et al. 2021). Another major shortcoming is the utilization of large amounts of NADs in the procedure to accomplish a realistic treatment period, thereby leading to the wastage of little prospective activity. Studies have been carried out to isolate powdery leftovers of the water treatment procedures by application of membranes which in turn enhance the price of the overall procedure. Some studies have conveyed the detrimental effects of nanomaterials because of the addition of materials in water for its decontamination. For example, chlorination of water to get rid of pathogens present in it leads to the formation of cancer-causing by-products. Furthermore, owing to the small size of nanoparticles, they can enter the lymph and blood via the epithelial and endothelial barriers and travel further into the brain, heart, liver, and other organs of human beings (Pandey et al. 2023). Hence, future studies are required with a focus on advanced comprehension and refinement in the utilization of NADs in water treatment.

## CONCLUSIONS

Amidst the variety of wastewater treatment techniques available to date, the adsorption technique is the utmost proficient and long-established one. The adsorption technique can exterminate organic as well as inorganic pollutants without creating any by-products or noxious intermediary substances. Consequently, it has an extensive pertinence in eradicating contaminants from a water resource. In recent times, NADs have been used in the adsorption process due to their exceptional assets. Thus, NADs are considered next-generation adsorbents and execute very well in restraining pollutants from wastewater. Various NADs can be utilized, like nanometals and their oxides and carbon nanotubes, because of the unique assets like a large surface expanse, stability, etc. However, there are still some shortcomings that limit the promotion of these materials. These shortcomings include the cost-effectiveness of the method, ecological apprehensions, and practical challenges like scaling up to the industrial level and system setup. Additionally, there are a few other challenges linked to the size of these materials, where the separation of nano adsorbents from water is a grave concern. Also, the availability of large quantities of nano adsorbents with low costs for water treatment destinations can be a serious issue for commercial procedures. Furthermore, preventing the release of used nanomaterials into the environment is a serious challenge because they accumulate for long periods.

In the future, novel studies can be performed with these materials in order to advance their applications in wastewater remediation, along with more investigations regarding the limitations of NADs.

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