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Adsorption and Removal of Pollutants (dyes) from Wastewater Using Different Types of Low-cost Adsorbents: A review

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ABSTRACT: Discharge of dye-containing wastewater from various industries like textile, leather, paint, cosmetics etc. industries contributes to large amounts of pollution, leading to serious environmental problems such as quality deterioration of fresh water and endangering ecosystems' health. One of the conventional anti-fungal agent in aquaculture which should be eliminated from wastewaters prior to releasing it into natural water resources. Tremendous volumes of colored wastewater are generated in textile, leather, paint, cosmetics etc. industries, causing eternal damage to the water resources. Wastewaters released from dye production and application industries are responsible for water pollution. Untreated disposal of the colored water into receiving water body causes damage to aquatic life and human bodies. Due to the high toxicity of dyes at low concentrations, they must be treated before being discharged into the receiving body of water. Therefore, environmental legislation has imposed severe limits on the discharged effluents' concentrations from dyestuff manufacturing and textile industries.

INTRODUCTION

A nanomaterial is defined as a material with any external dimensions in the nanoscale or having an internal structure or surface structure in the nanoscale – the length ranging from approximately 1 nm to 100 nm. Materials in this size range have different properties from their bulk state and exhibit size and morphology-dependent properties, making them interesting to researchers because of their myriads of application in different fields of study. Transition metal oxides are one among the different class of nanomaterial which are heavily studied. These oxides are of particular interest because of their varied metal to oxygen ratio and many phases with different structures, properties, and applications. Zinc oxide (ZnO), which belongs to II-VI transition metal oxides, is one of the most widely studied oxides as a result of its interesting physicochemical, optoelectronic properties and hence its application in many areas including in catalysis, sensors, medicine, personal care products, paints, batteries, solar cells, memory devices, electronic and spintronics, and in agriculture to control crop disease and as bio fertilizer[1-3] A detailed overview of the different areas of application of ZnO is given in reference [4, 5]. In particular, by virtue of its chemical stability, biocompatibility, and biodegradability nature, it has been extensively used as an antibacterial agent in antifouling application and in the radiation-assisted degradation of organic contaminants in wastewater treatment. Nowadays, access to clean water is becoming an increasing demand. However, due to the increase of urbanization and expansion of factories such as paper and ink, textile, leather, and others, the water body is becoming contaminated with

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residual waste discharged from these factories, causing harm to humans and aquatic lives.[6, 7].

Dyes

Currently, contamination of water is one of the major problems of the entire globe, due to the improper discharge of used water of the industries into the environment, high utilization of chemical fertilizers in agricultural fields, construction of roads, buildings, etc. [8-10]. Many chemical industries are dealing with the dyes, and among them, the large quantity of dye utilization and wastewater discharge after the process is being done by the textile industries exclusively. In the process of fiber conversion to yarn, yarn to fabric, dyeing and finishing, the textile industries use a large volume of water, numerous chemicals, auxiliary chemicals, dyes, and sizing materials [11]. The usage of such harmful materials has been resulting in water contamination and environmental pollution. The water released after the fabric preparation consists of dissolved solids, color, noxious metals (chromium), printing gums (pentachlorophenol, detergents), sequestering agents (trisodium polyphosphate and sodium hexametaphosphate, chlorine, azo dyes), and stain removers (CCl_4, residual chlorine, fixing agents like; formaldehyde and benzidine). Most of the aforementioned chemicals are harmful and a threat to the environment [12]. For the dyeing process, 60–70% of azo group dyes are used by most textile industries. Around 15–20% of the total dye is discharged into the environment during the process, which is dreadful harm to the environment [13-15]. The majority of the dyes used in this process are mutagenic, toxic, and recalcitrant to the microbial action's breakdown and favors the formation of the carcinogenic integrated under anaerobic degradation[16, 17].

Classification of dyes

Disperse dyes

The majority of disperse dyes are based on azo structures; however, violet and blue colors are often obtained from anthraquinones derivatives [18]. These dyes are frequently insoluble or sparingly soluble in water, non-ionic in character, and applied to hydrophobic fibers from an aqueous dispersion [19, 20]. They are predominantly used on polyester but have found application to nylon, cellulose acetate, and acrylic fibers; although some of the wet-fastness properties of the dyes on these substrates are poor, disperse azo dyes are the largest category among dispersing dyes that accounts for more than 50% of the total disperse dyes. These dyes are among the persistent class of dyes due to their recalcitrant nature and non-biodegradable behavior [21].

Direct dyes

The first direct dye was Congo red, discovered in 1884 [22, 23]. According to the same other, [24] has been confirmed, the direct or substantive dyes, which have a strong affinity for cellulose fibers, [25, 26]. They are mainly used for coloring paper products; these dyes have an appearance of solidity during washing [27] have been confirmed to, the direct dyes are classified according to many parameters such as chromophore, fastness properties, or application characteristics. The chromophoric Group of direct dyes includes azo, stilbene, oxazine, and phthalocyanine, with some thiazole and copper complex azo dyes [28, 29].

Reactive dyes

Reactive dyes have become very popular due to their high wet fastness, brilliance, and range of hues, [30], reported that reactive dyes are widely used because of the ability of their reactive groups to bind to the fibers, their stability, and their processing conditions, etc. [31]. These dyes are the second-largest classes of dyes [32, 33]. These dyes can form a covalent bond with the amine or sulfhydryl groups of proteins in textile fibers [34].

Vat dyes

Reported that vat dye is known for better color fastness, characteristically excellent light and wet fastness properties. They are mainly soluble in hot water, and some are soluble in the presence of little Na_2CO_3. This dye class are intended for application to cellulosic fibers because of their affinity for this last, and their application on nanofibers has not been investigated to date [35, 36]. Indigo belongs to the generic family of vat dyes. Its actual Colour Index number is C.I. Vat Blue 1, [37]. The most important natural vat dye is Indigo or indigotin found as its glucoside, indican, in various species of the indigo plant Indigofera. shows the examples of the vat dyes, [38]

Basic dyes

Reported that these dyes may have either the positive charge located on an ammonium group or the delocalized charge on the dye cation found in many triarylmethane, xanthene's and acridine dyes, [39-40] demonstrated that these dyes classes are applied with retarders because of poor migration properties at the boil,[21]. Reported that these dyes are usually applied to acrylic, paper, and nylon substrates but can also find use in some modified polyester substrates. The basic dyes contain a quaternary amine group which most often forms an integral part of the formula, but this is not systematic. Sometimes a positively charged oxygen or sulfur atom replaces nitrogen [23].

Acid dye

Reported that, acid dyes: As the name suggests, they are "acids", the molecule has one or more acid functions (SO_3H and COOH), [40]. Their acid nature explains their affinity for the basic functions of fibers, such as polyamides. As a representative element of this family of dyes, mention may be made of the red Congo, [31]. These dyes, especially for sulfonic acid dyes, are widely used in the textile, pharmaceutical, printing, leather, dye, paper, and other fields because of their bright colors, and high solubility, [14-16] confirmed that the acid dyes their usage constitutes about 30% 40% of the total consumption of dyes, and they are applied extensively on nylon, cotton, wool. They are usually applied at acidic pH. [15].

Sulfur dyes

The first Sulphur dye was prepared in 1873 by Croissant and Bretonniere, In 1966, sulfur dyes represented 9.1% of total US dye production and 15.8% of the dyes made for use on cellulosic fibers, and the world production was estimated at 110,000–120,000 tons per year, [32]. Sulfur dyes are a type of vat dye [25, 28] that demonstrated that sulfur dyes are high molecular weight dyes obtained by the sulfurization of organic compounds. These commonly used dyes to dye cellulose are converted into sodium-derived leuco by reduction using sodium sulfide [15]. They are applied to the fiber from a substantive leuco compound in the same way as for vat dyes [26].

Azo dyes

Azo dyes are the most used dyes. They are the main components of the textile industry used in the dyeing process [7]. They were reported to constitute 60–70% of all dyestuff concerning textile production [8]. Azo dyes belong to the largest family of textile dyes, comprising 60–70% of all textile dyes in practice [11]. The functional Group characterizes these dyes (-N = N-), uniting two symmetrical and/or asymmetrical identical or non-azo alkyl or aryl radicals [40]. They constitute the largest family, including 70% of the world's synthetic dyes' annual production [31]. In situ, azo colors are formed inside textiles by the reaction of two colorless or slightly colored compounds called naphthols, or C.I,[40]. Azo dyes containing heterocyclic rings lead to bright and deep shade [33]. The main disadvantage of azo dyes is to give dull shades on the blue-violet color range; between 60 and 70% of azo dyes are toxic, carcinogenic. They are resistant to biodegradability due to their chemical structure,[24].

MATERIAL AND METHODS

Methods of dye removal

Adsorption on low-cost bio sorbents

The adsorption process: Are one of the potential and efficient methods among all the possible techniques for colored effluent treatment due to its low initial investment, design simplicity, and availability of low-cost adsorbents [38]. Low-cost and readily available natural bio adsorbents are commonly applied for the elimination of several kinds of pollutants from wastewater. They are enriched with specific surface area and functional properties. Bio sorbents are considered a possible substitute for the costly and currently existing activated carbon for the uptake of the dyes from aqueous media [12-16].

Adsorption on activated carbon

Activated carbon (AC) is a special type of porous carbon material processed to have small, low-volume pores that enlarge the surface area. AC is suitable for the small molecules being used for the purification of gases and liquids. A variety of active carbons with different porosity can be obtained by controlling carbonization and activation. For removing IC, AC has been used extensively by researchers [3-7]. The charcoal was carbonized at different temperatures 800°C, 1000°C, and 1200°C. The method for producing great-capacity AC is not completely studied in emerging countries. Also, there are several problems with the regeneration of utilized AC. Nowadays, there is a countless interest in discovering cheap alternatives effective to the existing AC commercial [19-21]. Discovering effective and inexpensive AC might contribute to eco-friendly ability and offer benefits for future commercial applications. The prices of AC prepared from biomaterials are very little compared to the cost of commercial activated carbon. Waste materials that have been successfully utilized to production AC in the recent past contain bagasse, coir pith, orange peel, sunflower seed hull, coffee shell, waste wood, pine cone, coconut tree, oil palm shell, hazelnut husks, pine-fruit shell, corn cob, rice hulls, apricot stone and Coconut husk [24-26].

Adsorption on clay materials

The adsorbents developed from clay materials are characterized by large specific surface area, surface functionalities, and mechanical and chemical stabilities. These unique properties have established these materials as efficient adsorbents for removing different layers of layered double hydroxides (LDHs). These are lamellar inorganic solids with a brucite-like structure similar to hydrotalcite. Clays are defined as fine minerals grained that might plastic in-kind clays may be hardened when fired or dried and include suitable water contents. Clays mostly include phyllosilicates, yet the other contents find they might locate harden or either plasticity when dried or fried [29-32]. Clays can be distinguish from other soil fine-grained via their several in size, mineralogy [33] reported montmorillonite- sematic, bentonite, kaolinite, chlorite, and Elite the major kinds of clays. The Group of the Kaolinite clay contains the kaolinite mineral, halloysite, dickie, and nitrite. The group smectite contains pyrophyelite, vermiculite, talc, nontronite, saponite, sauconite and montmorillonite. The Group of the IL lite clay contains the clay micas. Elite is the only common mineral[35-38]. Chlorites are not constantly considered clay; some -times they are classified as groups separate inside the phyllosilicates. Naturally, Zeolites occurring minerals silicate, that container to be synthesized at commercial level. Possibly clinoptilolite is the utmost abundant of N40 natural species zeolite. The properties of the adsorption of zeolite clay depend upon their ion-exchange capabilities [40, 44].

Removal through electrochemical treatment

A viable alternative to biological treatment, electrochemical processes for abatement of pollution has been introduced in textile wastewater treatment. The merits and versatility of the electrochemical process are various and well documented. The electrode/catalyst is immobilized; hence, it is not necessary to separate the catalyst from the reaction mixture. It is very easy to do automation and control the variables such as the current and potential of the system[21-24].

Electrocoagulation

Due to its simple operation, high removal efficiency, fewer chemicals, and little sludge production, electrocoagulation is an attractive technology for wastewater treatment. Nowadays, electrocoagulation is getting much more attention as an alternative electrochemical treatment method in textile wastewater treatment research. Electrode configuration comprised of aluminum, mild steel, and aluminum/ mild steel pairs are employed to remove IC from the wastewater using the electrocoagulation technique [17].

Coupling process with electrochemical and activated carbon

Commercial Granular Activated Carbon (GAC) and Electrocoagulation (EC) are involved with the coupling process, which has been applied to separate dyes from aqueous solutions [20]. Photoelectrocatalytic technique, Integrated electrochemical application

Removal through chemisorption

In chemisorption's process, strong interactions, hydrogen bonds, covalent, and ionic bond formation happen between two atoms or molecules [27]. In this chemical reaction process, substantial sharing of electrons between the surface of adsorbent and adsorbate leads to covalent or ionic bond formation.

Removal by microbial biotransformation

The biomasses can decompose the organic and inorganic species like dyes, metals, and odor-causing materials. Fungi, algae, bacteria, sludge from biological wastewater treatment plants, by-products from fermentation factories, or seaweedsmay are sources for this biomass [28].

Removal by membrane filtration

The Nanofiber membrane is characterized by fine pores with high porosity and high specific surface area which is suitable for uptake of heavy metals and dyes from contaminated water. Generally, the electrospinning process is applied to prepare the Nanofiber membrane.

Removal by Nanocomposite materials

The mesoporous silica with the surface morphology-based materials is exhibited different types of dye removal according to the porosity of the material. The graphene oxide (GO)/polyaniline (PANI)/manganese oxide (Mn2O3) ternary Nanocomposites were successfully produced by using the one-pot method via in situ chemical oxidative polymerization of aniline in an acidic medium with MnO2 as an oxidant [19].

Photocatalytic degradation of dye

A material (catalyst) activated by adsorbing a photon and can accelerate a photo catalytic reaction without being consumed is known as a photo catalyst. These substances are invariably semiconductors. Semiconducting oxide-based photocatalysts have been progressively more used for environmental purification through solar energy conversion due to their high potentiality for wastewater treatment. This procedure is identified as an advanced oxidation process (AOP). After the development in the 1970s, heterogeneous photocatalytic oxidation has drawn significant attraction by researchers, and several studies have been performed on the application for the decompose of wastewater contaminants. Several semiconductors (TiO2, ZnO, Fe_2O_3, CdS, ZnS) were used as photocatalysts, but TiO2 was most frequently studied of its capability of breaking down organic pollutants. In the last few years, the AOP treatment using photocatalytic semiconductors has been applied for wastewater treatment, including indigo carmine dyes removal[20, 21,46].

RESULTS AND DISCUSSION

Nanotechnology

Nanotechnology is one of the new sciences that is used to provide a broad range of novel potential applications and increase promising technologies for newly future various applications. A unique part of nanotechnology is the "vastly increased ratio of surface area to volume," present in several Nanoscale materials, which begins different possibilities in surface-based sciences [32]. Nanoscale substances have the potential to advance the environment, both through the direct applications of these substances to detect, prevent, and eliminate pollutants, as well as indirectly by employing nanotechnology to design cleaner industrial processes and make environmentally responsible products [33]. Carbon nanotubes can be defined as materials that represent a connection to both graphite and fullerenes. The first experimental index of carbon nanotubes was reported in 1991 in multi-walled carbon nanotubes (MWCNTs). Carbon nanotubes have distinctive properties, such as electrical conductivity and high thermal, great strength, high stiffness, and specific adsorption properties. These materials can be used in a wide range of applications, such as removing organic and inorganic pollutants from wastewater. Another promising application of CNTs is their use as composite materials in many useful applications such as environmental, industrial, and photovoltaic applications[34].

Structure of Carbon Nanotubes

Carbon Nanotube is one-dimensional material. Mostly materials to result from nanotechnology, contain atoms are bonded that each other by building an sp2 hybridization in a graphite sheet or graphene Which is closed, which with honeycomb atomic arrangement making a network of hexagons. Carbon nanotube consists of carbon atoms structured in layers of graphene rolled into seamless cylinders [35]. A carbon nanotube can be arranged into two main structures: single-walled and multi-walled; the first type is a single-walled carbon nanotube (SWCNTs), a monolayer of carbon atoms in a graphene sheet rolled into a cylinder structure. Depending on the tube's diameter, folding angle, the direction of the wrap, and behave as helicity, nanotubes can behave metallic or semiconducting.. The second type is multi-walled carbon nanotubes (MWCNTs), which consist of many layers of graphene sheets rolled into a cylinder structure. The new type of CNTs is (FWCNTs) a few-walled carbon nanotube, a special type of MWCNT that consists of (two-six) layers of graphene sheets. They are considered as an extreme structure between SWCNTs and MWNTs. They have a diameter in the range of (0.3 -100) nm; the length may be reached several centimeters. The length of SWCNTs is recorded as more than 18 cm [26]. The diameter of (SWCNT) is about 1 nm while (MWCNTs). It reached 10 nm, while for few-walled carbon nanotubes (SWCNTs), the diameter is ranged (4-7) nm [14,45].

Classification of Carbon Nanotubes

According to many physical and chemical properties such as chirality, Conductivity, and number of graphene layers. Carbon nanotube can be classified.

Classification according to the Conductivity

According to the Conductivity, the carbon nanotube can be classified into two types, metallic and semiconducting. In general Conductivity of CNTs is affected by the chirality. The chirality's are determined by the(n,m) values of unit vectors.According to of these values, the carbon nanotube is two types: the first is when n-m is a multiple of 3 or an armchair, then the nanotube is metallic; the second refers to n-m not being a multiple of 3, or a zig-zag or chiral, then the nanotube is semiconductor [23-27].

Classification according to the layers

Carbon nanotubes can be classified depending on the number of layers of graphene into two types, as shown in Figure 1. **I-** Multi-walled carbon nanotubes (MWCNTs): This structure contains many layers of graphene sheets that are enfolded around to form it. **II-** Single-walled carbon nanotubes (SWCNTs): This class structure of nanotubes includes one layer of the graphene sheet. Recently, two classes have been added: they are double-walled carbon nanotubes (SWCNTs), which contain two layers of graphene, and few-walled carbon nanotubes (FWCNTs) , which contain (2-6) layers of graphene sheets [34, 46].

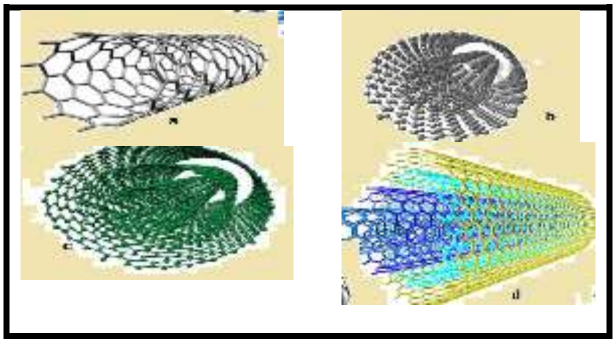

Figure 1. Schematic digram for the types of carbon nanotubes that are classified by the number of graphene layers (a) SWNT (b) DWNT (c) FWNT (d) MWNT.

C-Classification according to the Chirality

Chiral molecules are those which consist of different groups around the central atom that are non-superimposable on their mirror image. There are three special forms of carbon nanotubes, be determined by the wrapping of graphene sheets into the cylinder. Armchair, zigzag and chiral. The chiral of carbon nanotubes are uniquely characterized by the vector [31] . (Table1).

Table 1. Comparison of several Synthetic-Carbon Nanotube (CNT) for the adsorption of dyes.

Sorbent	Pollutant	E%	q_{max}(mg/g)	C_o (mg./l)	Dose (g/l)	t(h)	Ref.
CNT - Fe3O4	Methylene blue (MB)	98	248	50	0.2	3	[101]
MWCNTs/Fe3O4 (PDA + PEI)	Methylene blue (MB)	87	1449	200-700	0.75	2	[102]
MWCNTs/Fe3O4 (PDA + PEI)	Congo red (CR)	83	1006	200-700	0.68	2	[102]
MWCNTs/Fe3O4 (PDA + PEI)	Methyl orange (MO)	80	935	200-700	0.49	2	[102]
MWCNTs	Methylene blue (MB)	95	578	5-100	0.75	24	[103]
MWCNTs-COOH	Bromothymol blue	97	55	10-70	0.8	1	[104]
SWCNTs	Malachite green(MG)	88	26.68	30	0.01	1	[105]
SWCNTs	Methyl orange(MO)	77	26.16	30	0.01	1	[105]
CS/MWCNTs	Congo red(CR)	95	450.4	500	0. 2	24	[106]

Analytical techniques for CNTs

It is important to note that no single analytical technique is capable of defining the purity of a CNT sample. Several techniques have been used to characterize the structure and morphology of CNTs, to determine the purity of CNT materials; the most extensively used techniques: TEM, SEM, IR, Proton, Thermogravimetric analysis, and NMR Compared with analytical techniques to determine SWNT purity, microscopy techniques (TEM, SEM) seem to be the only ones that can confirm the presence of MWNTs in a sample. The most extensively used techniques.

Proton NMR has been used to monitor the progress of CNT functionalization. The presence of functional groups can be predicted by characteristic peaks arising from the magnetic environment's difference. 1H-NMR of functionalized CNTs is characterized by broad bands for protons close to the CNT, becoming sharper with distance. [33-38] .

IR spectroscopy is primarily a qualitative tool used to identify functional groups and the nature of their attachment to CNT sidewalls. Different functional groups absorb characteristic frequencies of IR radiation, giving rise to a fingerprint identification of bonds. It is a complementary technique to NMR to confirm the presence of bonds between CNTs and attached moieties [21].

TEM is used to determine the morphology and to give qualitative insight into the purity of produced CNTs. TEM uniquely provides qualitative information on the size, shape, and structure of carbonaceous materials, as well as non-CNT structured impurities in a sample. [10-13] However, it is unable to identify metallic impurities and does not differentiate SWNTs from MWNTs.

SEM is the only technique that can provide information on both CNT morphology [12] and the metallic impurity content. It is the most widely used technique to evaluate bulk productions of CNTs. In its conventional setting, the technique is limited by its inability to differentiate catalyst and carbonaceous impurities from CNTs[13]. However, the metallic content of CNT samples is routinely estimated by SEM coupled with an energy dispersive x-ray analysis detector (SEM-EDX) [14].

Thermogravimetric analysis is used to quantitatively determine the amount of carbon and noncarbon materials in bulk CNT samples, as well as CNT homogeneity and thermal stability. [13] This is a nonselective method for assessing CNT quality because it does not differentiate CNTs from metallic impurities present in the sample. It is therefore used in conjunction with other techniques. [18].

CONCLUSIONS

Nanotechnology is still in its very early stages. However, the numerous applications of this unique technology have garnered considerable interest in the medical community. The emergence of this technology may potentially revolutionize our approach to the central and peripheral nervous system pathologies. Nanotechnology has shown promise in its ability to construct complex nanoparticle scaffolds, create composites with various diagnostic and therapeutic utility materials, selectively deliver specific elements such as drugs to specific target tissues, and stimulate local cells to induce tissue regeneration. Compared with other pharmacologic agents and vectors, nanoparticles demonstrate an advantage in their physio-chemical-biological properties, including their extremely small size, polarity or electrical conductance, favorable shape, and greater surface area to volume ratio.

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not applied.

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