Nanocomposites for Dye Removal from Water

Mohamed A. Abdelwahab, Maie Mousa, and Nehal Salahuddin

Abstract The pollution of water by potentially dyes, is severe form of environmental impact. Traditional wastewater treatment are inadequate and cannot encounter the basic standards of water quality at sensible cost or processing time. Removal of dyes from aquatic surroundings has become a main alarm due to environmental problems and the possible hazards and hazards posed by them. Nowadays, the adsorption method as one of the most effective methods of eliminating pollutants has fascinated growing consideration among chemists and environmental researchers. However, one of the tasks is to design more effective adsorbents besides preparing them via greener and safer approaches. Nanocomposites are considered talented materials for the removal of potentially toxic dyes from aqueous solution through adsorption process. The present chapter deals with the utilization of nanocomposites for removal of dyes.

Keywords Dyes · Nanocomposites · Adsorpotion

1 Introduction

Nature has offered an abundance of resources for maintaining and grow life on the earth. Water is the essential source in the world, and today's world challenges serious issues in meeting the growing demand for safe drinking water, specifically in developing countries [97, 106]. A severe worry and vital goal for human life is clean drinking water. Moreover, water is a vital resource for food manufacturing and the preservation of human health as well as environmental protection. One of the most important challenges in the globe is the lack of clean drinking water [14, 35]. The dumping of industrial effluents into our waterways is a significant source of water contamination, which poses a severe environmental risk [68]. Drinking water can

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be contaminated from discharged agricultural, sewage and industrial waste, making it difficult to provide safe drinking water to nearby populations and ecosystems. According to statistical projections, approximately one billion persons living in dry countries will face clean water scarcity by 2025 [29]. Around 3.2 million person die every year as a result of a lack of clean water or sanitary conditions, particularly in poor nations [110]. Water is the most essential material for all species, notably humans, because water makes up around 60% of the human body. Contamination of the environment impacts the world's three essential components: air, soil and water. Due to population expansion, industrial and human activities have increased without equivalent environmental care, and hydric resources are being progressively depleted [80]. Water treatment is a theme of concern for our health and our environment. Drinking water sources must be treated to eliminate disease-causing chemicals.

1.1 Sources of Dyes and Its Classification

The scarcity of natural pigments and enhancements in dye manufacturing technology have made researchers and manufacturers progressively more dependent on synthetic dyes. Dyes are broadly utilized in productions, for example, paper, plastics, textiles and paints. Dyes can be categorized according to the source (natural and synthetic dyes), chemical structure (chromophores) (nitro and nitoso dyes, anthraquinone dyes, Azo dyes, triarylmethane dyes, phthalocyanine dyes, indigoid dyes and sulfur dyes) and application method (disperse dyes, acid and basic dyes, reactive dyes, vat dyes and direct dyes) as shown in Fig. 1 [15].

The amount of dyes engendered annually is assessed to be more than 700,000 tonnes, with 10–15% being released in wastewater [32, 60]. Synthetic dye present in the water is one of the most severe issues challenging environmental protection

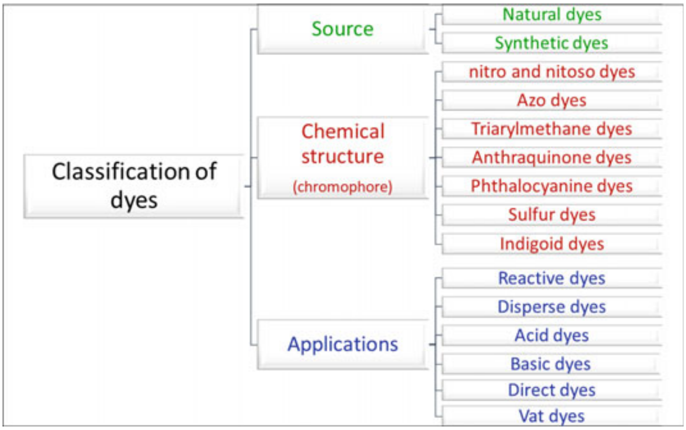

Fig. 1 Classification of dyes

strategies, owing to their difficulty being degraded into less dangerous compounds for human health and the environment. Dyes are harmful organic compounds with limited biodegradability that perform a substantial role in environmental issues such as eutrophication, visual pollution and water system disturbances [90]. Furthermore, the existence of dyes as pollutants in water resources can lower water quality and create carcinogenic and mutagenic consequences that can infect humans and wildlife. Dyes can affect a variety of difficulties when they infiltrate the environment. Because most of these organic materials are very stable toward microbial attacks, humidity, oxidizing agents and solar radiation if dismissed into rivers [11, 19], dyes can produce a decline in water properties by exposing color to the water and affecting the photosynthetic action of aquatic organisms by obstructing the access of solar light [51]. Recently, there has been much interest in eliminating coloured contaminants from wastewater [23].

1.2 Dye Removal Techniques

Nowadays, the most common processes in water treatment utilized by municipal water systems (mostly surface water treatment) involve coagulation and flocculation, sedimentation, filtration and disinfection [33]. Numerous methods have been utilized to eliminate dyes from contaminated media in both industrial wastewater and water resources such as electrochemical and membrane processes, biological treatment, coagulation, chemical oxidation, adsorption and aerobic microbial degradation (Fig. 2) [71, 89, 113]. Water treatment technology for heavy metals and dyes removal is dependent on the type of contaminant to be removed or basis for the treatment, whereas water treatment methods that are acceptable for dye removal may not be suitable for removing heavy metals. For example, chemical precipitation is used to remove heavy metals, although it is not proposed for colour removal from water. Chemical precipitation procedures are beneficial in the treatment of water and the creation of water treatment products and media [22, 55]. Treatment methods have been employed to focus on the elimination of recalcitrant dyes from wastewater and aquatic water. On the other hand, the adsorption method is widely used because of its several benefits, including economic feasibility, high efficiency in removing the dyes, and ease of processing as well as cost-effectiveness for dye removal from aquatic water and wastewater [20, 21]. The adsorption process includes a number of steps: (i) dissolving dyes in solution, (ii) exterior dye diffusion to the adsorbents' surrounds, (iii) intra/internal -particle diffusion (filling nanoparticle pores with dye materials), and (iv) desorption or adsorption on interior sites [3]. The strengths, weaknesses, opportunities and threats (SWOT) of persistent dye pollution removal based on adsorption techniques were effectively discussed on the next section [5].

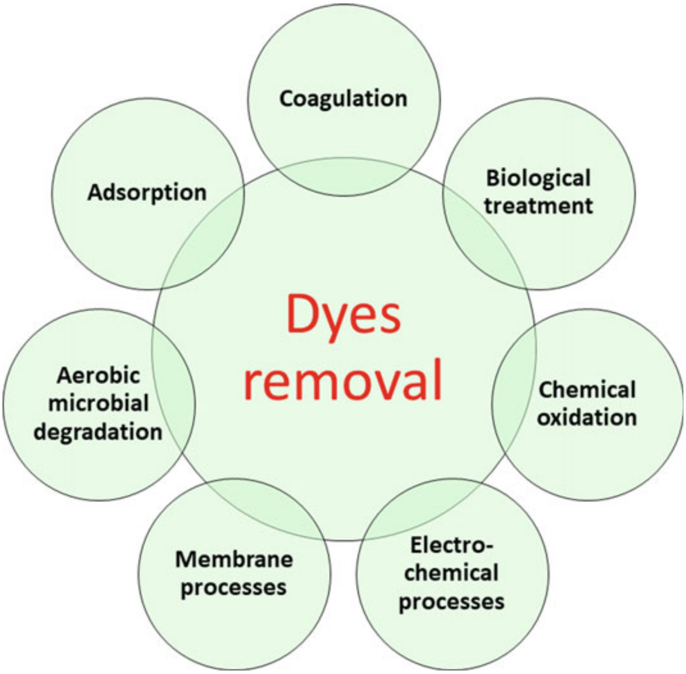

Fig. 2 Possible techniques used to remove dyes and water pollutants from contaminated media in both water resources and industrial wastewater

2 Nanocomposite Material

Nanocomposite material has expanded substantially to comprise a huge range of structures such as one, two and three-dimensional as well as amorphous materials, particularly prepared from various materials and combined together at the nanoscale. This fast-growing industry is producing a lot of new materials with unique features by mixing characteristics from different components/materials to produce a single material. There's also a chance that new qualities emerge that aren't present in the parent component materials.

2.1 Classification of Nanocomposites

In general, the organic–inorganic nanocomposite materials class are a rapidly expanding field of study. Effort is engrossed on the ability to get control of the

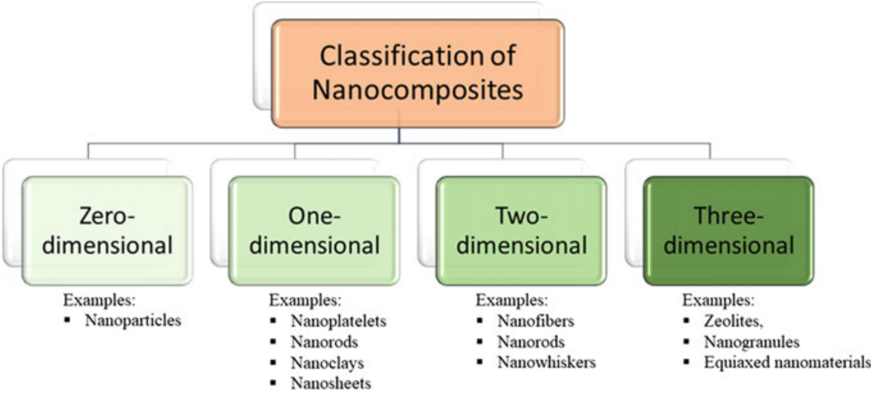

Fig. 3 Nanocomposites classification based on the dimensions

nanoscale structures via innovative synthetic approaches. The properties of nanocomposite materials depend not only on the characteristics of the matrix and the reinforcing agent/fillers, but also on their morphology and interfacial features. Nanocomposites could be classified according to the structure of nanofillers into four dimension (Fig. 3):

(a) Nanofillers with zero-dimension: Nano dimensions in any direction, for example, nanoparticles. They don't have any dimension exceed 100 nm. They might be of the amorphous, crystalline, metallic or ceramic nature.
(b) Nanofillers with one-dimension: Nano dimensions in only one direction, for example, nanotubes and nanowires (needle-type shape). Nanoclays, nanoplatelets, nanosheets and nanorods are an excellent examples of nanofillers with one-dimension.
(c) Nanofillers with two-dimension: Nano dimensions in two directions, for example, carbon nanotubes and silicates. It might also involve nanorods, nanowhiskers and nanofibers.
(d) Nanofillers with three-dimension: Nano dimensions in three directions, for example, nanogranules and zeolites.

2.2 *Efficiency of Nanocomposites for Dye Removal*

For dye removal, various organic and inorganic adsorbents (ex. Fly-ash, zeolite, saw-dust, mesoporous silica and activated carbon) are applied. These adsorbents have some drawbacks such as poor selectivity, low adsorption capacity and higher cost [78]. Adsorption using nanocomposites is the best way to remove the dye from wastewater due to its straightforward method, cheap method and efficacy to various kinds of dyes [28]. By engineering several functional groups (carboxylate, amino, acid, and hydroxyl groups) or the amalgamation of adsorbents, including metal oxides, graphene, graphene oxide (GO), and metal–organic frameworks (MOFs) in the nanocomposites, the separation capacity can be significantly improved [24, 73]. Because most dyes are resistant to biodegradation, photodegradation and oxidation, traditional biological and physico-chemical treatment techniques are unsuccessful for dye removal. Table 1 summarizes the removal efficacy and maximum adsorption capacity of different nanocomposites twards different types of dyes.

2.2.1 Clay-Based Nanocomposites for Dye Removal

Various inorganic clay has been used to remove the pollutions from the wastewater, such as laponite, sepiolite, attapulgite and montmorillonite [58]. Among these clay minerals, layered silicates montomorllonite (MMT) or nanoclay is an excellent reinforcing filler for removing the cationic and anionic dyes and can be used in different applications such as biomedical, agriculture, packaging and aerospace applications [1]. MMT has different characteristics such as high modulus, cheap material, high CEC (cation exchange capacity), non-toxicity, high surface area and ease recyclability [53]. MMT is an alumina octahedral silicates sandwiched among two tetrahedral silicate sheets [6]. By using cation exchange, the interlayer spacing of MMT was increased to enhance the adsorption capability of MMT minerals [1].

MMT has been applied in a different matrix for dye removals, such as polyvinyl alcohol and chitosan [36, 48]. The matrix of the nanocomposites also acting an crucial role in enhancing removal of the dyes. One of these matrices that was mixed with nanoclay is the hydrogel polymers (acrylic acid-acrylamide based polymers and/or copolymers) which have high adsorption capacity, oxygen barrier, thermal stability and flame retardant that can be applied in different applications (ex., agriculture and healthcare) [16, 54]. Another study utilized poly (acrylamide-co-sodium acrylate) in existence of biopolymer carrageenan and Na-MMT to enhance the absorbency of crystal violet dye (CV) from wastewater [58]. The best result has been obtained by using 10 wt.% of MMT due to enhancement of osmotic pressure. The water absorption decreased by an increasing amount of MMT, which works as a multifunctional crosslinker.

Kasgoz and Durmus [47] mixed MMT with acrylamide (AAm) and 2-acrylamido-2-methylpropanesulfonic acid sodium salt (AMPSNa) to eliminate two cationic dyes (Brilliant Cresyl Blue (BCB) and Safranine-T (ST)) from an aqueous solution. The authors found that the incorporation of MMT into the hydrogel improved the swelling capacity and diffusion capacity. However, these hydrogel composites have some drawback to the environment as the material can not be degraded which cause a "white pollution" [72].

Research is growing in the production of inexpensive, biodegradable, biocompatible and natural polymeric sorbents to overcome the disadvantages of using synthetic polymeric sorbent that are expensive and difficult to regenerate. A novel and green superabsorbent cellulose-clay nanocomposite hydrogels was produced by chemical cross-linking to remove the methylene blue dye (MB) [72]. The new materials displayed superior absorption capacity for MB solution (97% removal

Table 1 The removal efficacy and maximum adsorption capacity of different nanocomposites twards different types of dyes

Adsorbent code	The maximum removal efficiencies (%)	The maximun adsorption capacity (Q_{max})mg $g^{-1.}$	Type of Dye	References
Starch-g-PAAc/CNWs	90	2236	Methylene blue	Gomes et al. [30]
AAm-AMPSNa/clay hydrogel	–	526.3 500.0	Safranine-T Brilliant Cresyl Blue	Kaşgöz et al. [47]
GO-Cu-MOF	90	262	Methylene blue	Firouzjaei et al. [18]
G/Fe_3O_4	99.4	89.4	Fuchsine	Wang et al. [101]
Cellulose/MMT	98	1065	Methylene blue	Peng et al. [72] 2016
Chitosan/silica/ZnO	95	293.3	Methylene blue	Hassan et al [33]
Carbon/iron oxide	–	46.94	Rhodamine B	Singh et al. [95]
DTN	100	1,632	Direct blue 78	Hayati [34]
	94	1,220	Direct red 80	
Magnetic Fe@MgO		7299.2	Methyl orange	Ge et al. [26]
PES18P4CWNT0	98.7	–	Malachite green	Mousavi et al. [64]
Fe_3O_4@SiO_2-NH_2	97	–	Methyl red	Kamari et al. [44]
MCNC	99	–	Direct red16	Rafieian et al. [79]
MNC	94.21	223.82	Nylosan	Muntean et al. [65]
	85.31	114.68	Blue	
	96.50	286.91	Chromazurol S, Safranin T	

(continued)

Table 1 (continued)

Adsorbent code	The maximum removal efficiencies (%)	The maximun adsorption capacity (Q_{max})mg $g^{-1.}$	Type of Dye	References
A magnetic nanocomposite	–	113.31	Crystal violet	Singh et al. [96]
Nanocomposite superabsorbents	91.2	135.1	Crystal violet	Mahdavinia et al. [58]
CS/ZnO	96.7	–	Methylene blue	Mostafa et al. [63]
Zn0.5Co0.5Al0.5Fe1.46- La0.04O4	76	–		Ahmed et al. [7]
$CoFe_2O_4eSiO_2$	-	75.5	Malachite green	Amiri et al. [13]
ZIF-8@CNT	73.5	3300	Malachite green	Abdi et al. [2]
ZIF-8@GO	77	2034		
PANI-ZSP	–	12	Methylene blue	Gupta et al. [31]
Sm-MOF/GO	91	–	Rhodamine B	Yang et al. [108]
Chitosan/silk fibroin//hydroxyapatite	-	476	Methylene blue	Salama et al. [88]
Fe_3O_4@SiO_2-Zr-MOF	98	128	Methylene blue	Huang et al. [40]
	100	219	Methyl orange	
ZIF-67@Fe_3O_4@ESM	100	250.81	Basic Red 18	Mahmoodi et al. [59]
Ppy NF/Zn-Fe LDH	89.54	63.4	Safranin dye	Mohamed et al. [61]
G-SO_3H/Fe_3O_4	95.3	199.3	Methyl orange	Wang et al. [104]
	96.8	216.8	Brilliant yellow	
	95.5	200.6	Alizarin red	

(continued)

Table 1 (continued)

Adsorbent code	The maximum removal efficiencies (%)	The maximun adsorption capacity (Q_{max})mg $g^{-1.}$	Type of Dye	References
$Fe_3O_4 - TSC$	–	435	Malachite green	Alqadami et al. [11]
$MnFe_2O_4$/GO	94	46.08	Neutral red	Katubi et al. [45]
RHC-Fe_3O_4	–	321	Methylene Blue	Yang et al. [109]
PANI/NiO/MnO_2	97	248.4	Methyl orange	Abd Ali et al. [10]
BCA	99.5	142.8	RhodamineB	Mohammed et al. [62]
Zinc oxide	99	–	Initial dye	Nakkeeran et al. [66]
Graphene oxide intercalated montmorillonite	96	746.27	Crystal violet	Puri and Sumana [77]
(MZ/CS/AL)	–	6.14	Malachite green	Kazemiand Javanbakht [46]
PPy/Fe_3O_4/SiO_2	100	361.43	Congo red	Alzahrani et al. [12]
CuO/ZnO-T	80	-	Reactive yellow	Sharma et al. [94]
	86		Basic viole	
Cc-PEI	–	–	Methyl blue Rhodamine B	Liu et al. [57]
NiO-CMC	–	3.03	Methylene blue	Albokheet et al. [9]
(Fe_3O_4/ZA), Fe_3O_4	96.8	40.36	Methylene blue	Tran et al. [100]
$NiFe_2O_4$/GO (NGO)	–	476.19	Methylene blue	Thy et al. [99]

(continued)

Table 1 (continued)

Adsorbent code	The maximum removal efficiencies (%)	The maximun adsorption capacity (Q_{max})mg $g^{-1.}$	Type of Dye	References
CMC coated Fe_3O_4@SiO_2 MNPs	–	22.7	Methylene blue	Zirak et al. [114]
CMC/kC/AMMT	98	10.75	Methylene blue	Liu et al. [56]
CMC-AM/nHA-MS	–	–	Acid Blue 113	Varaprasad et al. (2018)
GONH	98.9	348.82	MV cationic dye	Bardajee et al. [82]
CS/CNFs	–	259.13	Cango red	Kafil et al. [43]
		553.12	Acid blue 1	
Montmorillonite-silica	84.10	172.40	Congo red	Zaccheus et al. [111]
NSG	99.5	38.46	Eriochrome Black T	Ahmed et al. [8]
Fe_3O_4@C	99.1	184	Rhodamine B	Kong et al. [49]
Z-Fe_3O_4 NC	97.5	2.57407	Methylene blue	Nyankson et al. [67]
St/Pn nanocomposite	99	3692.4	Reactive Black 5 Reactive Violet 4decolorized	Janaki et al. [41]
	98	422.40		
	87			
GO-$ZnFe_2O_4$	99	171	Methyl orange	Sadighian et al. [84]
XG-g-PAM/SiO_2	–	209.205	Congo red	Ghorai et al. [27]

(continued)

Table 1 (continued)

Adsorbent code	The maximum removal efficiencies (%)	The maximun adsorption capacity (Q_{max})mg $g^{-1.}$	Type of Dye	References
Guar gum-graft-poly (acrylamide)/silica (g-GG/SiO_2)	–	579.01	Reactive blue and Congo red	Pal et al. [69]
RGO–Ni)	90	65.31	Rhodamine B	Jinendra, U., et al. [42]
ZnS/NaAla-Gel-cl-polyAAm	97.37	-	Biebrich scarlet	Priya et al. [76]
	95.45		Crystal violet	
magnetic graphene oxide-hydrogels	–	769.23	Crystal violet	Pourjavadi et al. [74]
semi-interpenetrating network (semi-IPN)	97.58	–	Crystal violet	Sharma et al. [52]
	95.42		Eosin yellow	
XA-cl-poly(AAm)	96.82	4.7	Eosin yellow Eriochrome black-T	Sharma et al. [92]
	98.73	8.0		
BFO/Mrcl-poly(AAm)-IPN-poly(AA)	97.49	2.01	Auramine-O	Sharma et al. [93]
	95.39	3.06	Malachite green	
	94.56	7.55	Crystal violet	
PPy@CS/MMT/GO	99	76.4	Biebrich scarlet	Salahuddin et al. [87]
	97	26.7	Eosin Y	
	95		neutral red	
	66		Safranin O	
	85		Titan yellow	
PPy/CS/GO	63	0.12	Ponceau 4 R	Salahuddinet al. [86]
(M-PPy/CS/GO)	64.5	6.799	Ponceau 4 R	Salahuddin et al.[85]

efficacy for 10 mg.L^{-1} MB). Another study was applied for removing MB from wastewater by using carboxymethyl cellulose (CMC), biodegradable polysaccharide kappa-carrageenan (obtained from red seaweed) and activated MMT (A-MMT) [56]. A-MMT was prepared by an acid treatment process using sulfuric acid. The authors showed that the maximum MB removal was 98% (12.25 mg.g^{-1}) with using 0.4% A-MMT, and the adsorption still excellent after five consecutive cycles. Recently, multifunctional nanohybrid consisting of polypyrrole/chitosan, grapheneoxide and montmorillonite nanohybrid displayed high efficacy for the removal of neutral, anionic, and cationic dyes (Titan yellow, Safranin O, Neutral red, Eosiny, Biebrich scarlet) from aqueous solutio [87].

2.2.2 Carbon-Based Nanocomposites for Dye Removal

A novel adventure to improve the removal of the dye from aquatic media is directed to use carbon nano adsorbents graphene (discovered in 2004), which is an allotrope of carbon (SP^2 bonded carbon atom sheet) and has a two-dimensional hexagonal structure. Graphene powder has many different characteristics such as low density, superior surface area, chemically stable and superior thermal, electrical, mechanical and optical properties. Graphene has been used as an adsorbent material for removing the dye from aquatic water. Sulfonation favors the surface of the graphene Zeta potential, thus improve the interaction of sulfonated graphene with aromatic pollutants and thus promises the high adsorption of organic dyes on sulfonated graphene [25, 81, 91]. In addition, the sulfonated graphene nanosheets (G–SO_3H) have the ability to increase the dispersion in an aqueous solution [112].

Recently, blending graphene with a magnetic nanoparticles recorded unique chemical, physical characteristics and adsorption capacity compared to graphene material [4, 83]. Magnetic graphene nanocomposites have the ability to respond to a magnet, facilitating the separation of graphene from dispersion rapidly and effectively [25]. Wang et al. [101] found that a magnetic/graphene (Fe_3O_4/G) nanocomposite can effectively adsorb fuchsine dye. In another report, reduced GO-Nickel nanocomposite was prepared to remove Rhodamine B (RhB) dye from aquatic solution [42]. The results displayed that the pH influence the dye removal and showed higher removal at pH 8. According to the results of the kinetic investigation, RhB dye adsorption follows a pseudo-second-order kinetic model. Consequently, magnetic Fe_3O_4/G-SO_3H nanocomposite was synthesized and applied for removal of anionic dyes (methylene orange MO, alizarin red (AR) and brilliant yellow (BY)) and cationic dyes (victoria blue (VB), safranine T (ST) and neutral red (NR)) from aqueous medium as shown in Fig. 4a, b [105]. At pH 6, the new nanocomposite material showed higher adsorption capacity in cationic dyes than anionic dyes (Fig. 4c). The maximum adsorption capacities of G-SO_3H/Fe_3O_4 for ST, NR, and VB dyes were 199.3, 216.8, and 200.6 mg g^{-1} respectively [105].

Graphene oxide, an electron-rich, hydrophobic nanomaterial, with large specific area has been served as an amazingly brilliant adsorbent. Graphene oxide (GO)

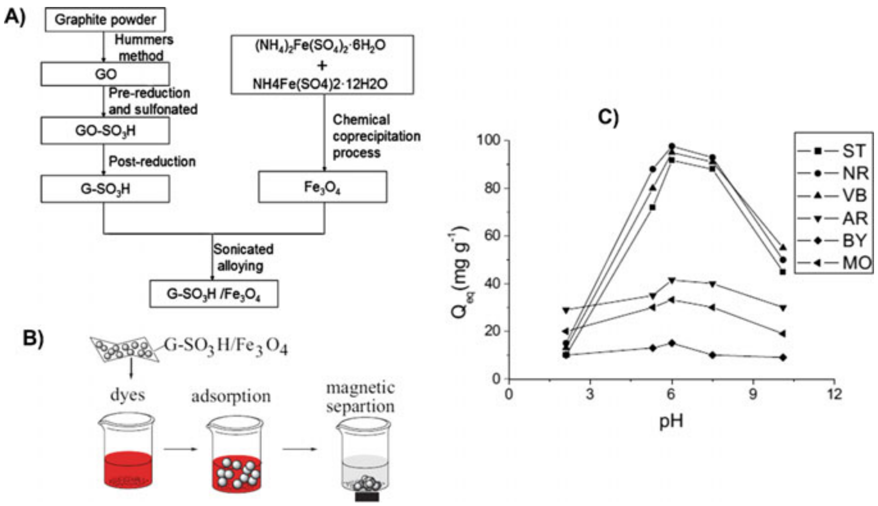

Fig. 4 **A** preparation of nanocomposite material from sulfonated graphene (G-SO_3H) and magnetic Fe_3O_4 nanomaterials; **B** Dye removal mechanism from aqueous solution using G- G-SO_3H/Fe_3O_4; **C** Effect of pH on the adsorption capacity of dye removal (MO, AR and BY) and cationic dyes (VB, ST and NR) (Reprinted with permission from Wang et al. [104])

is equivalent to graphene but with a high adsorption capacity and high surface-to-volume ratio and a functionalized group (hydroxyl, carboxyl and epoxy group) that allowed dispersion in the water and compatibility with hydrophilic dyes. Various researchers have explored the use of GO as an adsorbent for dye removals such as CV, MB, methylene orange and direct dye [23]. Puri and Sumana [77] formed a nanocomposite by the incorporation of GO into layered silicates MMT. The new nanocomposites have been used in the removal of CV dye from wastewater with a high adsorption capacity of ~746 mg/g as well as efficiently ~96%.

However, it was discovered that after the adsorption process, GO was very hard to eliminate from the aqueous medium by the standard separation method. As a result, it may raise the cost of industrial application while also polluting the treated water [17]. To solve the problem, GO hydrogel nanocomposites based on poly(acrylic acid)-g-salep polysaccharides biopolymer was applied for removing the cationic methyl violet (MV) dye in aqueous media [82]. By increasing the pH and temperature of the solution, the MV adsorption capacity on nanocomposites was enhanced, and adsorption equilibrium was achieved after 30 min.

In addition, it was reported that GO membranes are unstable in aqueous solutions due to poor adhesion between adjacent GO layers, limiting their application in wastewater. For this reason, a new nanocomposites membrane composed of samarium metal–organic framework (MOF) and GO was synthesized [108]. The Sm-MOF/GO (mass ratio of MOF: mass ratio of the reactant = 0.31) nanocomposite showed higher permeance (26 m^{-2} h^{-1} bar^{-1}) and high rejections (~91%) to MB and RhB. A porous crystalline 3D MOFs was made by coordination bonds from metal ions and organic ligands. Firouzjaei et al. observed the adsorption of MB by using copper-based MOF, immobilized on GO and reported that the adsorption capacity improved from 106 to 142 mg/g by increasing the temperature from 25 to 65 °C. Existence of GO in the MOF network enhanced the adsorption capability of capturing the dye [18]. Another MOF zeolitic imidazolate framework (ZIF-8) was incorporated with CNT and GO to prepare a new nanocomposite [2]. The new material was used to remove a malachite green (MG) dye from aquatic medium. Hybrid nanocomposite material showed a higher rate than sole MOF. The ZIF-GO nanocomposite material exhibited a higher adsorption capacity (3300 mg/g) than ZIF-8-CNT nanocomposites (2034 mg/g) at 20 °C.

A novel nanocomposite based on Chitosan/carbon nanoflowers was applied for adsorption of acid black1 (AB1) and cango red (CR) anionic dyes. The maximum adsorption was recorded to be 259.13 and 553.12 mg g^{-1}, respectively within 60 min [43]. In another report, the adsorption behavior of polypyrrole/chitosan/graphene oxide to Ponceau 4R (P4R) dye under different conditions such as different, salt concentrations, temperatures degrees and dye concentrations was studied and recorded high efficiency than the constituents [86].

It is worth to mention that magnetic adsorbents are characterized by high specific surface area, simple modification, structural flexibility, processing viability, mechanical strength properties and by applying an external magnetic field the adsorbents are easy recovered. The magnetic adsorbents comprise elements with magnetic properties. A new magnetic manganese ferrite ($MnFe_2SO_4$) nanoparticles was applied as magnetic support (synthesized by sol–gel route) for different adsorbents because of high magnetic and thermal characteristics, cheaper material, non-toxicity and high chemical stability. Thus, to enhance the adsorption of graphene, $MnFe_2SO_4$ was added as a support for reduced graphene adsorbent. The new nanomaterials showed high adsorption capacity to remove cationic dyes (MG and MB dye) [4]. Another study presented that GO/$MnFe_2SO_4$ has a high adsorption capacity to remove arsenic As^{5+}, As^{3+} and lead Pb^{2+} from aqueous water [50]. Katubi et al. (2021) displayed that GO/$MnFe_2O_4$ nanocomposite was efficient to remove NR dye with adsorption capacity of 46.08 mg/g. In another report, Zinc ferrite ($ZnFe_2O_4$) magnetic nanoparticles were used with GO in the removal of MO dye [84]. As well, Nickel ferrite ($NiFe_2O_4$) was utilized with GO to remove MB from an aquatic water [98]. Using 500 ppm MB at pH 10 within 150 min, the adsorption capacity achieved 343.98 mg/g.

2.2.3 Metal Oxide-Based Nanocomposites for Dye Removal

Metal oxide-based nanomaterials such as TiO_2, SiO_2, iron oxide, zinc oxide (ZnO), gallium oxide (Ga_2O_3), nickel oxide (NiO), copper oxide (CuO) [75] having exceptional functionality in their probable application to the detoxification of industrial effluents, groundwater, surface water and drinking water. However, limitations due to lose their selectivity, mechanical strength, and high capacity [70] were cured by immobilization these metal oxides into polymeric materials and designing core shell nanocomposites [38]. Mesoporous silica-gibbsite nanocomposites (NSG)

(62.34 $m^2 \cdot g^{-1}$, a pore radius of 22.717 nm, and a pore volume of 0.7081 $cm^3 \cdot g^{-1}$) recorded an efficient capacity to adsorb Eriochrome Black T dye (EBT) through an exothermic process (Inas A [8]. Magnetic nanocomposite based on cobalt ferrite-silica ($CoFe_2O_4eSiO_2$) recorded adsorbtion capacity of 75.5 ± 1.21 mg g^{-1} for the removal of MG dye from water. Interestinly the adsorbent was regenerated and used repeatedly and separated from water by applying a magnetic field [13].

Iron oxide (Fe_3O_4) nanoparticles have been used in designing nanocomposites due to their excellent properties such as high adsorption, superparamagnetic behavior, good compatibility, low toxicity, high surface energy, and large surface area [37, 102, 103]. The magnetic nanocomposite based on carbon/ iron oxide was applied for removal RhB from aqueous solution with maximum adsorption efficiency of 46.94 mg/g[95]. A shell of Fe_3O_4 MNP coated by a functionalized amorphous silica extracted from rice husk showed high efficiency for the removal of methyl red (MR) dye (97%) due to the presence of hydrophilic functional groups on the surface [44]. In another report, a magnetic Fe@MgO nanocomposites presented admirable adsorption properties towards methyl orange dye (MO) with the maximum adsorption capacities of 6947.9 mg•g^{-1} through chemical adsorption via complexing with MgO, associated with a slight mineralization of MO [26]. Consequently, magnetic nanoparticles dispersed within a matrix of activated carbon (MNC) recorded the maximum adsorption capacities of 223.82 mg g^{-1} for Nylosan Blue (NB), 114.68 mg g^{-1} for Chromazurol (ChS), and 286.91 mg g^{-1} for Basic Red 2 (BR2). The adsorption of three dyes on MNC was favorable, spontaneous, and endothermic with good efficiency (greater than 65%) afterward seven adsorption—desorption cycles [65]. The efficacy of a non magnetic carbon was compared with magnetic carbon nanocomposite for adsorption of CV dye from aquous solution. The adsorption capacity was improved from 40 mg/g to 81.7 upon using magnetic nanocomposite. The maximum adsorption was recorded to be 113.31 mg/g under the optimum conditions (concentration 240 mg/l; temperature 50 °C; pH 8.50; dose 1 g/l), which was very close to the experimental value (111.80 mg/g) [96].

A nano-sized $Fe_3O_4@SiO_2$ core (15 nm) coated with a shell of Zr-MOFs (about 5 nm) recorded high adsorbtion capacity for organic dyes [39] and remained unchanged after 6 recycles. In another report, zeolitic imidazolate framework-67 (ZIF-67) (1403.7 m^2/g) grown on the surface of Fe_3O_4@ egg shell membrane (1263.9 m^2 /g) recorded a maximum adsorption capacity of 250.81 mg/g for BR18 (Basic Red 18) following pseudo-second order [59]. Consequently, a core shell nanocomposite designed from $Zn_{0.5}Co_{0.5}$ $Al_{0.5}Fe_{1.46}La_{0.04}O_4$/PVP succeeded in purifying inked-water and dyes with higher efficiency of 90% in comparsion with pure ferrite (76%) (MA [7]. Trisodium citrate based magnetite nanocomposite (Fe_3O_4 −TSC) was applied for MG dye removal from aqueous medium [11]. The maximum adsorption capacity (qe) was calculated to be 435 mg g^{-1} following pseudo-second-order kinetic model with greatest recovery of MG dye in 0.1 M HCl. In another publication, magnetic Fe_3O_4/zeolite NaA nanocomposite exhibited [100] a high adsorption capacity (~40.36 mg · g^{-1}) and removal efficiency (~96.8%) compared with the zeolite NaA (~32.99 mg · g^{-1} and 79.11%, respectively) due to the interaction between active sites on the surfaces and edges of the invert spinel ferrite Fe_3O_4 nanoparticles and zeolite NaA with MB molecules.

It is wotarth to mention that multifunctional magnetite-polypyrrole/chitosan /grapheneoxide exhibited high adsorption efficiency of P4R dye contaminated water with easily magnetic separation. Moreover, it has been shown to be effective against antimicrobials (E. coli) and antifungals (Fusarium fungi). The adsorption kinetics of P4R dye follows the Langmuir isotherm equation and the pseudo- second-order kinetic model and the adsorption process was an endothermic (Nehal A [85]. In another report, modification of an Fe_3O_4/SiO_2 magnetic composite with polypyrrole (PPy) (PPy/Fe_3O_4/SiO_2) improved the maximum adsorption of (361 mg/g) CR dye with high ability to reuse the nanocomposite several times leading to reduction the overall cost of the treatment [112].

A bionanocomposite of sodium alginate, chitosan (MZ/CS/AL), infused with natural zeolite, and cross-linked with glutaraldehyde and $CaCl_2$ was used to remove methylene blue from aqueous solution [46] with high efficiency. Carboxymethyl cellulose (CMC) coated $Fe_3O_4@SiO_2$ core–shell magnetic nanoparticles (MNPs) exhibited an efficient adsorbent for the removal of cationic dye, due to the abundant adsorption site, easy separation using an external magnetic field and low cost [114].

Nano-hydroxyapatite polymeric hydrogels (Varaprasad et al., 2018) developed using waste shells (Clam and Magellan shell) carboxymethyl cellulose/acrylamide were investigated as a sorbent for Acid Blue 113 (AB) from aqueous AB solution. The diffusion coefficient value was increased with the increasing nano-hydroxyapatite content in the CMC-AM/nHA-CS (0.22353–0.27681 $cm^2\ s^{-1}$) and CMC-AM/nHA-MS (0.22377–0.29737 $cm^2\ s^{-1}$) hydrogels. Combination of conducting polymers e.g. polypyrrole with LDHs improved both the adsorption of cationic dyes as well as anionic dyes through modification of surface [61, 107].

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