

Nanomaterials and their derivative biocomposites for dye adsorption

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19.1 Introduction

Healthy water has been serving as the cornerstone for the sustenance of well-being and socioeconomic development of all forms of life on Earth, including human beings since the dawn of creation (Ahmed et al., 2019; Guo & Yu, 2021; Meehan et al., 2020; Rajasulochana & Preethy, 2016; Rojas & Horcajada, 2020; Safajou, Khojasteh, Salavati-Niasari, & Mortazavi-Derazkola, 2017) and its essential role in supporting the ecosystem, and the main triangulated freshwater competitors are irrefutable (Baig, Al-Zahrani, Schneider, Straquadine, & Mourad, 2019; Dong et al., 2013; Khoso, Wagan, Tunio, & Ansari, 2015; Ma et al., 2020; Pahl-Wostl et al., 2013; Panhwar et al., 2022).

However, in an epoch marked by a complex interplay of rapid population growth, industrialization, and urbanization, the matter of aquatic pollution has intensified to unprecedented levels and has become an alarming global concern, casting a shadow over the health of ecosystems and the well-being of plants and animals globally (Abebe, Murthy, Zereffa, & Qiang, 2020; Ahmed et al., 2019; Bijekar et al., 2022; Roy & Shamim, 2020; Ukaogo, Ewuzie, & Onwuka, 2020). As projected, by 2050, the global water demand will surge by 67%–134% (Hejazi et al., 2014) that will make the severe blow of water scarcity hit about 4.8–5.7 billion populace (Veldkamp et al., 2017) to the extent that this population will not have access to the 2–4 L/day drinking water needed by each person, talk less of about 200 L for hygiene and other needs (Panhwar et al., 2022).

Notably, this burgeoning crisis has been established to be fueled by the discharge of various effluents into the aquatic bodies with dye effluent coming to the fore from various industries shown in Fig. 19.1 (Karim, Dhar, & Hossain, 2018; Katheresan, Kansedo, & Lau, 2018; Priya & Selvan, 2017; Samsami, Mohamadi, Sarrafzadeh, Rene, & Firoozbahr, 2020; Solayman et al., 2023; Yaseen & Scholz, 2019). These dye pollutants not only mutilate the esthetic appeal of water resources but also pose grave threats to aquatic biotas and human health (Aslam et al., 2023; Raza et al., 2023; Srinivasan, Bankole, & Sadasivam, 2022). They are also complex (Emmanuel & Adesibikan, 2021), neurotoxic (Al-Gheethi et al., 2022; Kaur, Kaur, & Kaur, 2023; Köktürk, 2022; Verma & Gupta, 2022), and carcinogenic (Garg & Roy, 2022; Ma et al., 2022; Preethi et al., 2023; Sankar Sana, Haldhar, Parameswaranpillai, Chavali, & Kim, 2022) and impede the passage of sunlight into the water, which would impede photosynthesis and heighten the need for biological oxygen (Bal & Thakur, 2022; Lord, Neve, Keating, & Budhathoki-Uprety, 2022; Madima, Kefeni, Mishra, Mishra, & Kuvarega, 2022), therefore inhibiting the development of photoautotrophic organisms, endangering aquatic creatures, and escalating the scarcity and difficulty of access to clean water for eco-fundamental networking. Furthermore, the ramifications of these dye effluents are far-reaching, negatively impacting aquatic biodiversity, food chains, the delicate balance of the planet's biodiversity, and even human livelihoods (Emmanuel & Adesibikan, 2021; Emmanuel, Adesibikan, & Saliu, 2023; Emmanuel, Adesibikan, Saliu, & Opatola, 2023; Emmanuel, Olawoyin, Adesibikan, & Opatola, 2023).

Consequently, It is therefore of the highest necessity to develop effective and sustainable techniques for the treatment or removal of dye pollution (Bhatti et al., 2020; El-taweel et al., 2023) and for donkey year's years now, concerted effort in employing conventional treatment methods, such as adsorption (Saxena, Sharma, & Saxena, 2020),

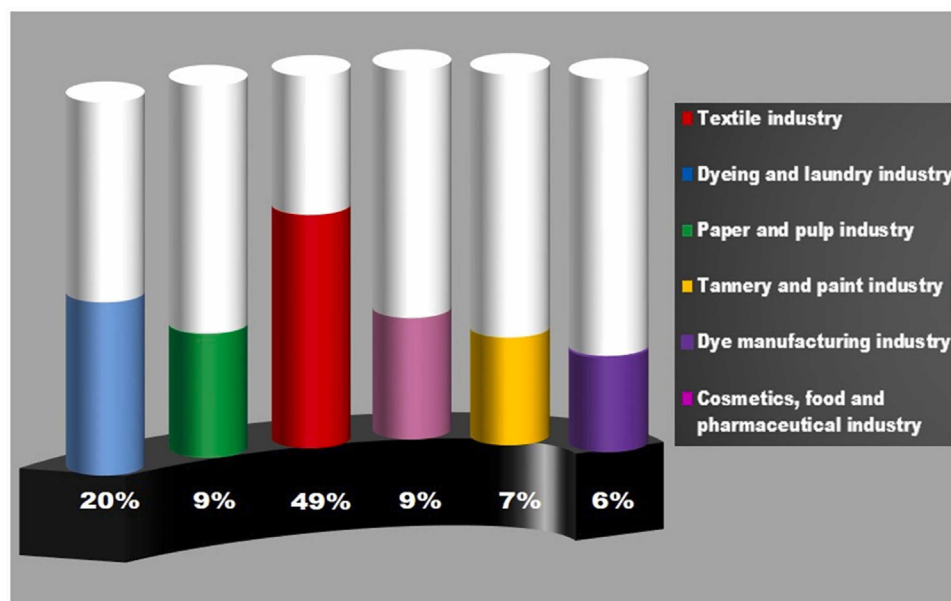


FIGURE 19.1 Sources of notorious dye runoffs. From Emmanuel, S. S., Adesibikan, A. A., Saliu, O. D., & Opatola, E. A. (2023). Greenly biosynthesized bimetallic nanoparticles for ecofriendly degradation of notorious dye pollutants: A review. *Plant Nano Biology*, 3. <https://doi.org/10.1016/j.plana.2023.100024>.

coagulation, flocculation (Abbas, Mohamed, Al-Sahari, Al-Gheethi, & Daud, 2021; Jin, Nan, Chen, Song, & Wu, 2023; Korpe, Venkateswara, & Rao, 2022; Tang, Shixin Zhang, & Zheng, 2021), reverse osmosis (Boudissa et al., 2019), micro/ultra-filtration (Eguagie, Hung, & Paul, 2021), and precipitation (Aragaw, Bogale, & Aragaw, 2021; Garg & Roy, 2022), has been made. Sadly, aside from adsorption, these other techniques have exhibited drawbacks as per efficiency and cost-effectiveness (Aragaw et al., 2021; Iqbal et al., 2021).

Interestingly, adsorption has been used with cost-effectiveness adsorbing materials such as activated carbon, biochar, clay, and zeolites, which has not given the scientific community the kind of ultra-efficiency, high selectivity, and regeneration desired (Aragaw et al., 2021; El-sayed, 2020), and this has prompted researchers to explore advanced techniques, leading to the emergence of nanomaterial-based adsorption as a promising avenue for ultra-efficient dye removal (Adel, Ahmed, Elabiad, & Mohamed, 2022; Amin, Alazba, & Manzoor, 2014; El-sayed, 2020; Jiang et al., 2023; Saxena et al., 2020; Uddin & Baig, 2019). Without gainsaying, the integration of nanomaterials has revolutionized this age-old principle (Saxena, Saxena, & Lochab, 2020). Specifically, this innovative approach capitalizes on the unparalleled properties of nanomaterials to boost the adsorption capacity, selectivity, and recyclability of adsorbents (Al Naim & El-Shamy, 2021; Du et al., 2020; El-sayed, 2020; Kumari et al., 2020; Xue et al., 2021). More specifically, nanoparticles, by their unusually high surface area-to-volume ratio, give an enlarged surface area for interaction with dye molecules, resulting in increased adsorption performance (Abebe, Alamirew, Whitehead, Charles, & Alemayehu, 2023; El-sayed, 2020). Even at low concentrations of dye pollutants, nanoparticles (NP's) tiny size allows them access to a greater number of active sites, leading to increased adsorption efficiencies (Abouzeid, Khiari, El-Wakil, & Dufresne, 2019; Adel et al., 2022; Tan et al., 2015; Zhang, Wang, Zhang, & Zhou, 2018). Furthermore, because of their adaptable surface qualities, which may be altered to adjust their affinity toward certain dye contaminants, the adsorption process is more selective (Awad et al., 2020; Safajou et al., 2017).

Notably, a remarkable development in this revolutionized nanomaterial-based adsorption process employing nanomaterial biocomposites instead of only nanomaterial came to the limelight in recent years. This adsorbing material coalesces the advantages of nanoparticles with that of natural materials such as agro-waste (Barasarathi, Abdullah, & Uche, 2022; Iqbal et al., 2021; Kaur, 2024; Nguyen et al., 2021), zeolite (Majid, AbdulRazak, & Noori, 2019; Piri, Mollahosseini, Khadir, & Hosseini, 2019), biopolymers (Abdulhameed, Jawad, & Mohammad, 2019; El-taweel et al., 2023), clay (e.g., hydroxyapatite, rectorite, kaolinite, and montmorillonite) (Abdullah, Shameli, Abdullah, & Abdullah, 2019), and geopolymers (Al-husseiny & Ebrahim, 2022; Falah, MacKenzie, Knibbe, Page, & Hanna, 2016; Maiti, Sarker, Maiti, Malik, & Xu, 2020).

In addition to enhancing nanomaterials' stability, strength, conductivity, and reusability, biocomposites also provide biodegradability and nontoxicity, answering queries associated with environmental impact (Iqbal et al., 2021; Maiti et al., 2020). These composites offer a dual-layered approach, with nanomaterials affording the adsorption capacity,

while biocomposites bestow structural integrity and compatibility with biological and aquatic systems (Emmanuel, Adesibikan, et al., 2023; Kaur, 2024).

On this note, this chapter delves into the captivating realm of dye pollutant adsorption using nanomaterials and their derivative biocomposites. It aims to provide a comprehensive overview of the recent advancements in this innovative approach to shed light on the synergetic strides made in employing a diverse range of emerging nanomaterial biocomposites and the mechanisms (including surface chemistry) underpinning their adsorption capabilities. Moreover, by synthesizing and analyzing the existing body of knowledge, this chapter seeks to empirically elucidate the key factors governing the adsorption process, such as adsorbent architectural makeup/properties, dye properties, and the influence of pH, temperature, initial dye concentration, contact time, and background electrolytes to unravel the intricacies of achieving optimal adsorption efficiency. In the end, this work was laced with challenges and frontier sections that can serve as future research hotspots for the scientific community. We believe that as environmental sustainability takes center stage, the insights garnered from this review work could help broad readerships, researchers, and industrialists who are interested in fighting for water security and ecological preservation and pave the way for the development of efficient, eco-friendly, and economically viable solutions to combat other deadly pollutants in our water resources as well.

19.2 Adsorption as a process for dye removal

The process of adsorption plays a crucial role in removing dye from contaminated water. It involves the attachment of dye molecules to a solid surface, either through chemical or physical interactions (Bayode et al., 2020). This process can occur on various adsorbent materials, such as activated carbon, silica gel, and zeolites. During adsorption, the dye molecules are attracted to the adsorbent material and become attached to its surface. As a result, the concentration of dye in the water decreases, leading to decolorization (Adeola, Abiodun, Adenuga, & Nomngongo, 2022; Adewuyi, 2022). This process is widely used in wastewater treatment plants and is an effective method for removing dyes from industrial effluents (Adewuyi, 2022). The adsorbent is the solid surface, while the dye adsorbed from the solution is the adsorbate. The two categories of adsorption are based on the nature of the interaction between the adsorbate and the adsorbent. When the interaction is physical, it is called physisorption, which is characterized by weak interaction forces like van der Waals forces. Conversely, when the interaction between the adsorbate and adsorbent is chemical, it is called chemisorption, where the interaction forces arise from chemical bonding (Bayode et al., 2020). Desorption is the opposite of adsorption, where adsorbate is removed from the adsorbent surface. In physisorption, weak forces make desorption easy, but it is difficult in chemisorption due to strong forces (Bayode et al., 2020).

19.2.1 Physicochemical properties of nanocomposite as dye adsorbent

Nanocomposites, composed of nanoscale components, show exceptional promise as dye adsorbents due to their unique physicochemical properties. By integrating nanoparticles into a composite structure, the material's adsorption capacity, selectivity, and overall efficacy for dye removal are significantly enhanced.

Nanocomposites possess high surface areas and porosities, primarily attributed to the nanoscale features of their components. This characteristic significantly increases the available sites for dye molecules to adsorb, leading to superior adsorption capacity. Additionally, the specific arrangement and morphology of nanoscale components in the composite structure play a vital role in dye adsorption. For instance, the presence of nanosheets, nanotubes, or nanoparticles with well-defined shapes and structures can significantly improve the accessibility of adsorption sites and provide a higher surface area for interactions with dye molecules. The chemical composition of nanocomposites can be tailored to optimize dye adsorption. Surface functionalization of nanoparticles or the incorporation of specific functional groups can enhance the affinity for certain types of dyes through chemical interactions such as hydrogen bonding or electrostatic forces. Furthermore, the surface charge of nanocomposites influences the electrostatic interactions between the adsorbent and dye molecules. By controlling the surface charge through modifications or choice of components, it is possible to selectively target and adsorb dyes with opposite charges. Nanocomposites' mechanical strength and stability are critical for their practical application in water treatment processes. Enhanced structural integrity ensures that the material can withstand physical stresses and maintain its adsorption efficiency over multiple cycles. The ability to regenerate and reuse the adsorbent is crucial for economic and environmental sustainability. Nanocomposites often exhibit excellent regeneration potential, allowing for the desorption of adsorbed dyes and the reuse of the material for multiple adsorption cycles. The selectivity of nanocomposites for specific dyes can be influenced by factors such as size, shape, and chemical properties. By tailoring the composition and structure of nanocomposites, it is possible to

design materials with high selectivity for target dyes. The temperature sensitivity of nanocomposites can also affect the adsorption process. Some materials exhibit temperature-dependent adsorption behaviors, which can be exploited for controlled and efficient dye removal under specific conditions.

19.2.2 Characteristics of dye pollutants affecting sorption by nanomaterials

The adsorption behavior of nanomaterials and their derivative biocomposites for various dye contaminants may vary greatly owing to the large disparities in their architectural functional group, bonds, and other properties (Jiang et al., 2023). Majorly, the characteristics of dye pollutants, such as aromaticity, molecular size, hydrophobicity, and polarity, are important factors affecting their interaction with nanomaterials (Cheng, Zhao, & Han, 2018; Jiang et al., 2023).

For instance, Cheng's research team (Cheng et al., 2018) reported that under the same experimental condition, more methyl orange (MO) was adsorbed by ZnO/ γ -Al₂O₃ NPs than methylene blue (MB) and rhodamine B (RhB). It was accentuated that the nanomaterial has a +ve-polarity, resulting in better adsorption for dye pollutants (like MO) with a -ve-polarity than the dye pollutants (like MB and RhB) with a +ve-polarity (Cheng et al., 2018). Other studies (Zhou et al., 2017) have confirmed that polarity is a primary factor in governing the adsorption of dye pollutants onto nanomaterials and their derivatives. Findings suggest that nanomaterials that have a +ve-polarity afford better adsorption efficiency for dye pollutants with a -ve-polarity and otherwise. Furthermore, as per hydrophobicity, the binding of the dye by polycations may be significantly influenced by hydrophobic contributions (Constantin et al., 2013). The adsorption rate of strongly hydrophobic dye and organic pollutants by adsorbents is slow (Jiang et al., 2023).

As for the molecular size, dye pollutants with different molecular sizes have different effective contact and interception effects on nanomaterials/nanocomposite, and thus the mechanism of adsorption and adsorption effect is also different (Anirudhan & Ramachandran, 2015; Jiang et al., 2023). Generally, dye and organic compounds with larger molecular sizes are hardly absorbed by adsorbents owing to the size exclusion effect and pore-filling mechanism, which saw to their restricted from entering pores smaller than their size (Han et al., 2014; Jiang et al., 2023; Zhu, Liu, Jiang, Lin, & Yang, 2022). For example, MB has the smallest size compared to RhB > MO, and it is much easier for it to enter the porous tunnels of the nanomaterials during adsorption operation and get trapped (Cheng et al., 2018). The above assertion was consistent with that of Dogan's team (Dogan, Satilmis, & Uyar, 2019). Also, a similar adsorption efficiency trend [adsorption capacity of polymer nanofibers material was 1000 mg/g for Sunset yellow (M.W: 452.38 g/mol), 454.55 mg/g for amaranth (M.W: 604.47 g/mol), and 344.83 mg/g for Fast Green (M.W: 808.85 g/mol) was reported by Thamer, Aldalbahi, Meera Moydeen, Rahaman, & El-Newehy (2021)].

Briefly, regarding the aromatic profile of dye pollutants, aromatic organic compounds can form π - π interactions with nanomaterials/nanobiocomposite and be tightly adsorbed during the adsorption process (Jiang et al., 2023; Zeng et al., 2019), and this interaction becomes appreciable with aromatic rings numerical strength of the irrespective dye pollutants. For instance, acridine orange (AO) and MO contain a naphthalene ring and a benzene ring, respectively, with the same ionic group. However, AO contains three aromatic rings compared to MO, and this makes the adsorption capacity of AO onto nanomaterial microspheres higher than that of MO, signifying that the bulkier the aromatic substituent the greater the binding force of the dye (Constantin et al., 2013). Nevertheless, the negative effects of some of these properties can be managed to some extent by manipulating the adsorbent architectural surface profile and adsorption parametric conditions such as pH and temperature that shall be discussed in another section of this chapter.

19.2.3 Effect of operating parameters

By and large, as seen in the foregoing section, most nanocomposite adsorbents have high adsorption capacity for dye pollutants. However, the efficiency of these adsorbents toward the adsorption of dyes varies because the adsorption operation is governed by different parametric variables (pH, temperature, contact time, initial concentration, background ions, and ionic strength), especially in real-life scenarios owing to the fact the architectural properties of adsorbent and dye vary and also background electrolytes in the solution can interact with the adsorbent surface (Umeh et al., 2023). The impact of these parameters on the elimination of dyes is expounded in the following paragraphs.

19.2.3.1 Effects of pH

pH is the negative log to base 10 of hydrogen ions (H⁺) or hydroxonium ion (H₃O⁺) concentration in each solution. This shows an imperative part in adsorption because elimination efficacy in this technique is dependent on the pH of the medium since its alteration tends to affect the level of melting properties (Soltani, Faramarzi, & Mousavi Parsa, 2021). The change in pH may also affect the charges on either the surface of the dye or material, thereby influencing

the amount of adsorption (Nayeri & Mousavi, 2020). In other words, pH alteration results initiate physical and chemical imbalances and produce different isomers with numerous active sites for adherence to the adsorbent surface (Panda et al., 2021). The impact of pH on any adsorption process is essentially described via the concept of the point of zero charge (pHpzc) (Umeh et al., 2023), which corresponds to the point where the electrokinetic behavior of the adsorbent is initiated. pHpzc refers to the point where the collective charges on the adsorbent surface correspond to zero. It is observed that at pH less than the pHpzc value, the vacant sites on the adsorbents' surface are positively charged, but at pH higher than the pHpzc, active surface sites on the adsorbent are negatively charged (Parimelazhagan, Natarajan, Shanbhag, Madivada, & Kumar, 2023). This implies that when the pH is greater than pHpzc, there is a favorable adsorption of anionic species (dyes) on any adsorbent and vice versa. Therefore pH directly affects the rate of adsorption; hence the evaluation of the starting pH of the dye is pivotal in any study (Amdeha, 2023). The main factors influencing the pH-dependent adsorption of dyes by nanomaterials are the surface chemistry of nanomaterials (Alswieleh, 2022; Osagie et al., 2021; Sadegh et al., 2017) and charge density (Homaeigohar, 2020; Talbot et al., 2021).

Parimelazhagan and his coworkers biosynthesized, Zn(OH)₂ nanomaterial using *Calotropis gigantea* (CG-Zn(OH)₂NPs) and employed the green NPs for the elimination of Coomassie violet and investigation of the effect of pH on its removal. The pHpzc was found to be 8.5, indicating that at a pH lower than the pHpzc the active sites of the adsorbent will be positively charged. But at elevated pH, the exterior of the adsorbent will be negatively charged, owing to the deprotonation of active binding sites with the solution's OH⁻ ions. The highest removal efficiency of 94.3% could occur at a pH higher than the pHpzc (Parimelazhagan et al., 2023).

In a study to examine the elimination of double positive dyes (MB and RhB) in water, Brennan and fellow researchers prepared four nanomaterials; graphene oxide (GO), F-GO, IC-rGO, and G that were used for the study. They observed that GO adsorbed MB more favorably at pH 6 with an elimination efficacy of 99.0% at an original dye amount of 160 mg/L, while a similar result was observed for F-GO but with a decreased removal efficiency of 96.1% at an original amount of 120 mg/L. However, lesser removal efficiency was observed for IC-rGO and G with 47.6% and 15.6%, respectively, at a starting dye amount of less than 40 mg/L. The authors did not report a detailed report on the influence of pH that probably would have improved the removal efficiency of the other materials (Mao, Sidhureddy, Thiruppathi, Wood, & Chen, 2020).

Fe₂O₃ nanoparticles were incorporated into ash (nFe-A) from agricultural waste leaves obtained from *Rosa canina* by Agarwal et al. and investigated the elimination of malachite green dye and evaluated the effects of pH on the adsorption efficiency. The authors reiterated that adsorption efficacy improved meaningfully when the pH of the dye solution increased from 3.0 to 8.0 from the range of 73.13%–78.26% resulting from the fact that the number of positively active sites on the adsorbent increased with the decrease in pH of solution. This resulted in the protonation of the carboxylic group on malachite green (MG), giving rise to a high density of positive charge at a smaller pH value resulting in electrostatic repulsion among the like charges on the adsorbate as well as nanomaterial, nFe-A. But at higher pH, the exterior of the nanomaterial turned negatively charged that enhances the elimination efficiency of malachite green due to electrostatic attraction (Agarwal, Tyagi, Gupta, Mashhadi, & Ghasemi, 2016). Other investigations on the impact of pH adsorption efficacy of dyes using nanoparticles are summarized in Table 19.1.

19.2.3.2 Effects of contact time

Interaction time is a crucial feature that affects the removal efficiency of dyes by nanomaterials using the adsorption technique. This is defined as the period in which the adsorbent (nanomaterials) are in contact with the adsorbate (dye molecules) or the period within which diffusion and adhesion of dye molecules into the nanomaterial take place in a given medium. Since adsorption could either be chemisorption or physisorption involving interaction, the longer the nanomaterials and dye molecules are in contact with each other, the better their interaction with each other and possibly more adsorption. But sometimes improving the interaction time results in to decline in elimination efficiency, perhaps owing to the exhaustion of the vacant binding spots available on the nanomaterial. In most scenarios, adsorption efficiency occurs in two phases, which accelerates within the first period of contact, but the removal becomes gradual in the second-period phase (Singh, Kumar, & Dwivedi, 2022). To explore the impact of interaction time on the elimination efficiency of nanomaterials toward dye removal by adsorption, several adsorption studies have reported the effects of contact time.

For instance, the research group of Joshi and colleagues explored the utilization of Fe₃O₄ doped activated carbon nanomaterial toward the elimination of methyl blue and brilliant green dyes in water and evaluated the impact of the interaction period from 0 to 120 minutes. They observed fast improvement in the adsorption efficiency within the first phase (20 minutes) owing to the accessibility of a free dynamic site on the surface of the nanomaterial for the

TABLE 19.1 Influence of pH and interaction period on the uptake efficiency/capacity of nanomaterials on dye elimination in aqueous solutions.

Nanomaterial	Dye adsorbed	Optimum pH	Optimum contact time (min)	Adsorption capacity or efficiency	References
CaFe ₂ O ₄	Reactive orange 12	2.0	40	276.92 mg/g 77%	Das & Debnath (2021)
Titanium dioxide	Reactive black 5	6.0	10	36.67 mg/ g	Shaheed and Hussein (2014)
Activated carbon@Fe ₃ O ₄	Methylene blue, malachite green, and Congo red	2.0 9.0 2.0	120	83.9% 94.3% 93.4%	Sivaprakash, Satheeshkumar and Krishna (2017)
Nanocellulose/chitosan/ <i>N,N'</i> -methylenebisacrylamide	Malachite green	6.0	30	42.0 mg/g	Goswami, Mishra, Prasad, and Bhatt (2022)
γ-Fe ₂ O ₃	Alizarin	11.0	60	23.2 mg/g	Badran and Khalaf (2020)
ZnO NPs- <i>Artocarpus heterophyllus</i>	Congo red	9.0	60	90%	Vidya, Manjunatha, Chandraprabha, Megha Rajshekar, & Antony Raj (2017)
FeNPs- <i>A. terreus</i> GS28	Congo red Direct blue-1	4–8 4.0–7.0	60	98% (48.7 mg/g) 79% (38.0 mg/g)	Singh et al. (2022)
ZnO NPs- <i>Ocimum sanctum</i>	Congo red	4.0	30	97%	Nayak, Sahoo, & Sahoo (2022)
Fe NPs- <i>Terminalia bellirica</i> plant extract	Congo red	–	–	86%	Jegadeesan, Gautham, Amirthavarshini, & Divya (2019) Gunarani (2019)
Fe ₃ O ₄ @AC	Methylene blue Brilliant green	Above 7.8	20	138 mg/g 166.6 mg/g	Joshi et al. (2019)
ZnO NPs- <i>Hibiscus rosasinen</i>	Congo red	4.0	20	95.5%	Debnath & Mondal (2020)
Sawdust/MnFe ₂ O ₄	Indigo Carmine	2.0	15	93%	Hashemian & Hidarian (2014)
Fe ₃ O ₄ @MIL-100(Fe)	Methyl red	4.0	360	586.34 mg/g	Dadfarnia, Shabani, Moradi, & Emami (2015)
Chitosan/Al ₂ O ₃ /magnetic iron oxide	Methyl orange	4–10	10	99%	Tanhaei, Ayati, Lahtinen, & Sillanpää (2015)
Chitosan-functionalized graphene oxide	Methylene blue	7.0	35	23.3 mg/g	Nayl et al. (2022)
GO reduced graphene oxide (RGO) M-RGO-N	Methylene blue	8.0	120 60 120	39.8 mg/g 36.6 mg/g 49.4 mg/g	Vassileva et al. (2023)
GO	Methylene blue	8.0	15	429.5	Soudagar, Akash, Venkat, Poiba, & Vangalapati (2022)
GO-CS	Methylene blue	5.0	125	7.5	Khiam et al. (2022)
Alumina	Orange G	2.5	5.0	98.4%	Banerjee et al. (2019)

adsorption of the dyes. Slight removal was detected after that until equilibrium was attained after 120 minutes, suggesting the unavailability of active sites for adherence of the dye particles due to occupation or exhaustion (Joshi, Garg, Kataria, & Kadirvelu, 2019).

Another study by Kurniawati and fellow investigators evaluated the effect of the interaction period on the elimination of RhB, methyl orange, and methyl blue adopting nanomaterials prepared from *langsat* shells. They observed that the highest adsorption capability of RhB occurred at the first interaction period of 60 minutes having a value of 11,578 mg/g, after which it decreased gradually. For methyl orange, the best interaction period was 90 minutes with the removal capability of 38,425 mg/g, representing the balance point between adsorption and desorption. After this, there was a decrease owing to the exhaustion of the available binding spots for further uptake of methyl orange by the material. Furthermore, for the attraction of methyl blue by the synthesized nanomaterial, there was an increase in uptake with a removal capacity of 36,735 mg/g within the contact time of 30–150 minutes, but from 180 minutes there was a gradual decrease in the uptake, corroborating the statement of occurrence of adsorption in two stages (Kurniawati, Bahrizal Sari, Adella, & Sy, 2021).

Einafshar's research group synthesized a multifunctional nanocomposite material combining graphene sheet, TiO₂, Ag/Al₂O₃, and β-cyclodextrin to form Ag/Al₂O₃/TiO₂@β-cyclodextrin-GO (AATG) adsorbent that were employed toward the elimination of MB, while the impact of interaction time was evaluated. From the result, a fast elimination efficacy of more than 99% was obtained within the first 5 minutes, and equilibrium was achieved at about 30 minutes by AATG, which was 20% greater than achieved by GO. This occurred because uptake on GO emanated due to its bidimensional structure, while that of AATG occurred on both surfaces of the two components of the synthesized nanomaterial (Einafshar, Amiri Farmad, Moshirian Farahi, & Einafshar, 2023).

In a research by Modwi and fellow investigators, they examined the elimination of RhB and evaluated the impact of equilibrium contact time utilizing a coupled nanohybrid adsorbent prepared by a simple method. The investigators observed for all the concentrations examined for 120 minutes, adsorption capacity surged significantly at the first 40 minutes of interaction time and then attained equilibrium. After that, there was a gradual decrease in the adsorption capacity, signifying a great influence on the adsorption of RhB using the synthesized nanocomposite material (Modwi et al., 2022).

Likewise, in another study for the elimination of MB via hydroxyapatite (HA)/gold (Au) nanocomposites biosynthesized using plant extract of *Acrocarpus fraxinifolius*, the impact of the interaction period toward the uptake of the dye was also evaluated. From the result obtained, it was shown that at a lower amount of 30 mg/L, a stable interaction of 90 minutes was attained with higher adsorption capacity. Still, at greater concentrations, equilibrium was achieved at a faster contact time but with lower adsorption capacity (Sharma et al., 2021).

19.2.3.3 Effects of initial dye concentration

The effect of the starting amount of dye is a vital physicochemical factor impacting the adsorption technique, as it could positively or negatively influence the adsorption efficiency via an upsurge or decline in the vacant energetic binding spots on the nanomaterial. In any sorption procedure, the effect of this factor on the efficacy of dye removal and adsorption capacity is determined as follows:

$$E\% = \frac{C_i - C_f}{C_i} \times 10 \quad (19.1)$$

$$q = ((C_i - C_f) \times V) / m \quad (19.2)$$

where E is the removal efficacy, q is the maximum uptake capacity (mg/g), C_i is the original dye amount (mg/L), C_f is the finishing dye amount (mg/L), m is the quantity of the nanomaterial (g), and V is the quantity of the solution (mL).

From Eqs. (19.1) and (19.2), it can be deduced that the elimination efficiency of dyes in solutions decreases as the initial concentration increases, it can also increase as the starting amount surges, while in some scenarios, no observable change (Rápó & Tonk, 2017). This implies in some cases that the greater the adsorbing species in the medium, the better the potentiality of the uptake, which in general is owing to the accessibility of excess binding spots for the dye molecules. In another perspective, every adsorbent possesses a specific number of dynamic sites on their surface; therefore an upsurge in the starting amount of the dye will improve the removal efficiency at a rapid rate with fast equilibrium attainment between adsorption and desorption. This results in to decline in the efficacy of the adsorption with no further adsorption (Panda et al., 2021).

Hambisa's team explored the influence of starting dye amount toward uptake efficacy/adsorption capacity of methyl orange dye from solution utilizing agricultural waste-based anchote peel nanomaterial (Hambisa, Regasa, Ejigu, & Senbeto, 2023). The author observed an improvement in uptake capacity with an upsurge in the starting dye amount until it got to 70 mg/L, and subsequent decline that could be attributed to presumably saturation or unavailability of active binding spots on the exterior part of the nanomaterial.

In another study, Kandisa's research group evaluated the elimination efficiency of MB dye by adopting a cheap adsorbent synthesized from the *Vigna trilobata* pod and determined the influence of the original dye amount on the efficacy. The authors observed that the quantity of dye eliminated depended on the starting amount of the dye in aqueous media, with increased removal efficacy attained at lower dye concentrations. A decrease in efficiency was observed from 94.5% to 90.5%, and the adsorption capability at the stability point was improved from 8.4 to 34.2 mg/g with upsurge in starting concentrations from 25 to 150 mg/L. This implies that the accessibility of energetic binding sites at the beginning became rarer with the rise in the concentration of the dye solution (Kandisa, Narayana Saibaba, Shaik, & Gopinadh, 2021).

Vassileva and his team investigated the elimination efficacy of MB utilizing three graphene-based nanomaterials and evaluated the conditions influencing the optimum attainment of adsorption capacity, including initial dye concentration. From the result obtained the proposed all three nanomaterials with high adsorption ability for MB, but at equilibrium, M-RGO-N produced the highest decline in the initial concentration of MB (Vassileva et al., 2023).

Another research group of Torkian and Gholinezhad synthesized a nanomaterial via modification of mesoporous carbon (CMK-3) with Fe_3O_4 and studied the impact initial amount of the dye that was aimed at the elimination of malachite green dye from water. From the experiment, they suggested that an influence of starting malachite green amount increased adsorption capacity of the two nanomaterials (CMK-3 and Fe-CMK-3) to 307.78 and 300.58 mg/L, respectively, starting from 20.91 to 20.21. This was suggested to result from the upsurge in the motivating potency of the concentration rises with a greater starting amount that rapidly attains equilibrium at a concentration below 600 mg/L, indicating higher adsorption capacity for lower concentrations due to available active binding sites on the nanomaterials (Torkian & Gholinezhad, 2015). Other investigations on the impacts of starting concentrations on the uptake of dyes using nanomaterials are presented in Table 19.2.

19.2.3.4 Effects of competitive adsorption and ionic strength effect

In the adsorption technique, competition and ionic strength play key parts in defining the efficacy of an adsorption process in the removal of dyes using nanomaterials owing to their unique and efficient surface morphology of the nanomaterials. Competitive adsorption refers to the situation where there are multiple adsorbates or interference within a given matrix that struggle for the accessible active binding spots on an adsorbent/nanomaterial. The presence of this competition can allow a great reduction in the adsorption efficiency/capability of the nanomaterial as it tends to be occupied first by the adsorbates that possess a high affinity for the available active sites. Therefore in carrying adsorption, the presence of other organic or inorganic species that can adhere to the nanomaterials other than the dye molecules may tend to decrease the efficiency of the material toward the targeted dye. For instance, in a study with two basic dyes, blue 41 and yellow 28 in a dual arrangement adopting two different adsorbents compared with a single system, basic blue 41 was favored because it has a greater affinity to the pore of the adsorbent compared to basic yellow 28 (Regti, El Kassimi, Laamari, & El Haddad, 2017). Parisi and his research fellows evaluated the competition between metal ions and dye molecules in a waste matrix via the adsorption process by utilizing montmorillonite clay mineral nanomaterial adsorbent. They observed that there was no uptake of metal ions in acidic environments; however, the dyes adsorbed under acidic conditions (Parisi et al., 2019).

Single and competitive adsorption studies were also carried out by Yunusa and his colleagues toward the sequestration of crystal violet and malachite green, cationic dyes using activated carbon nanoparticles obtained from *Balanites aegyptiaca* seed shell. They observed that single adsorption showed great adsorption capacities for both dyes at pH 8 and an equilibrium contact period of 50 and 60 minutes for malachite green and crystal violet, respectively. The lower adsorption capacities could be attributed to the antagonistic effect resulting from their vying for similar binding sites (Yunusa et al., 2021).

However, ionic strength is the level of concentration of some ion in the same matrix as the adsorbate that can modify the adsorption process via an adjustment of the interaction among the adsorbent and the adsorbate using different physicochemical processes such as electrostatic repulsion, ion exchange, and steric hindrance. All these mechanisms tend to reduce the adsorption performance of the nanomaterials for dyes when available in a given matrix by decreasing the availability of active binding sites for adsorption. It has been established that real-life water matrix is contaminated

TABLE 19.2 Effects of initial concentration on the adsorption capacity of nanomaterials for elimination of dyes.

Nanomaterial	Dye adsorbed	Initial concentration range (mg/L)	Equilibrium time (min)	Adsorption capacity or efficiency	References
MgTiO ₃ @g-C ₃ N ₄	Rhodamine B	5.0–100.0	–	18.5–215 mg/g	Modwi et al. (2022)
ZnO@zeolite	Methylene blue	1.0–7.0	35	67.8%–94.8%	Elfeky, Youssef, & Elzaref (2020)
Fe ₃ O ₄ @alginate beads	Methylene blue	20.0–300.0	60	106.38 mg/g	Rezaei, Haghshenasfard, & Moheb (2017)
Polyvinylpyrrolidone (PVP)-supported AgNPs-coated AC PVP-supported AuNPs-coated AC	Congo red	2–10	30 and 270	75.0%–71.0% 88.0%–85.0%	(Pal & Deb, 2014)
Modified multiwalled carbon nanotube	Basic orange and methyl violet	5.0–30	20 30	55%–32.6% 74%–65.7%	Mohammed, Razak, & Hussein (2014)
Cobalt ferrite	Congo red	10–100	130	97.84%	Sidhaarth, Jeyanthi, Baskar, & Vinod Kumar (2018)
Nanobentonite MgO-impregnated clay	Malachite green	50–250	60	94.5%–86% 92%–83%	Hussien Hamad (2023)
Silver@Ca(OH) ₂	Basic green 4	10–100	25	99.9%	Sheeja, Sampath, & Manivel (2019)
NiO	Reactive black 5	25–2000	20	25–1489.79 mg/g 95.15% to 76.93%	GONEN & BİÇER (2022)
CA@Fe ₃ O ₄	Crystal violet	0.5–20	40	48.26% to 26.45%	Jangra et al. (2023)
Biogenic silver	Crystal violet	500–1500	20	704.7 mg/g	Moghadas et al. (2020)
Green nickle oxide	Methylene blue and methyl orange	25 20	100 160	96% 88%	Rashid, Salman, Mohammed, & Mahdi (2022)
Zirconium oxide	Acid violet 19	10–100	150	31 mg/g	E.Marghany, M.Moustafa, El-Sharkawy, Abdel-Azem, & A.Ali (2022)
Zinc oxide	Methylene blue	20–80	60	96.25%	Ahmad & Igwegbe (2020)
Fe ₃ O ₄ /C	Methylene blue	20–100	60	169.533 mg/g and 208.33 mg/g	Khoshsang, Ghaffarinejad, Kazemi, & Jabarian (2018)
Zeolite	Congo red	25–30	100	219.2 mg/g and 289.4 mg/g	Imessaoudene et al. (2023)
Sansevieria plant leaves extract titanium dioxide	Congo red	10–50	60	93.92–56.79%	Salim & AL-Mammar (2023)
Green (red algae <i>A. plumula</i> extract) zinc oxide	Direct blue 106	10–40	120	370.37 mg/g	Eleryan et al. (2023)

with different organic and inorganic ions that impede the adsorption efficiency; hence the effect of coexisting anions at different concentrations was studied by Banerjee and team using NO_3^- , Cl^- , CO_3^{2-} , SO_4^{2-} , PO_4^{3-} , and $\text{C}_2\text{O}_4^{2-}$ with orange, green (OG) in aqueous solution. They observed that NO_3^- , Cl^- , and CO_3^{2-} ions in the availability of the dye showed no significant impact on the uptake efficiency, but SO_4^{2-} , PO_4^{3-} , and $\text{C}_2\text{O}_4^{2-}$ anions showed a remarkable impact on the uptake efficacy of OG. When the amount of PO_4^{3-} and $\text{C}_2\text{O}_4^{2-}$ were faintly upsurged, the adsorption capacity dropped down to 30 mg/g from 93.3 mg/g (98.4%). They attributed the high affinity of the SO_4^{2-} ion to the production of complexes on metal hydroxide surfaces (Banerjee, Dubey, Gautam, Chattopadhyaya, & Sharma, 2019).

Chen and team synthesized organosilica nanoparticles using an essential 2-degree amine that was utilized toward adsorption of phenol red dye in water and examined the influence of ionic strength utilizing NaCl of different concentrations (0.0–4.0 M), mixed with 4 mg/L of the dye. Adsorption efficacy was to have reduced rapidly as the ionic strength rose from 0.0 to 0.5 mol/dm³, followed by a gradual decrease as the ionic strength increased more to 4 M, suggesting the uptake process to be from electrostatic attractions (Chen et al., 2017).

In the uptake of Congo red adopting nanofibers of polyacrylonitrile doped with iron(III) oxide nanoparticles ($\text{Fe}_3\text{O}_4\text{NPs}$), the effects of ionic strength were also evaluated using NaCl with a concentration range of 0.1–0.4 M and 30 mg/L concentration of potassium chloride (KCl). The authors observed that the hydrophobicity improved when there was an upsurge in the amount of ions, which decreased the solubility in the aqueous solution resulting in to decrease in adsorption capacity (Jadoo & Naser, 2020). In a similar study for the elimination of MB utilizing green synthesized nanoparticles from coffee (MNLC) and peanut husk (MNLPH) improved with Fe_3O_4 , the effect of ionic strength was studied using sodium chloride with varying concentrations of 0%–10% (w/v). They observed that a rise in the amount of NaCl caused a reduction in the adsorption efficiency for MB using MNLC but increased for concentrations of up to 8%–10% (w/v) using MNLPH (Besharati, Alizadeh, & Shariati, 2018). In general, increase in concentration of competing ionic species would tend to decrease the adsorption capacities of nanomaterials toward dye molecules due to few available vacant sites for adsorption.

19.2.3.5 Effect of temperature and adsorption spontaneity

The impact of temperature on the uptake of dyes using nanomaterials is also a vital factor as it influences the removal method by adjusting the reaction to either endothermic or exothermic and vice versa (Yeow, Wong, & Hadibarata, 2021). The influence of temperature on the adsorption method results in alteration in spontaneity and physicochemical interactive activity occurring between adsorbent and dye molecules leading to either increase or decrease in adsorption efficiency concerning temperature. During an adsorption process, if the reduction in adsorption rate results from an upsurge in temperature, the mechanism is exothermic, while the reverse is endothermic (Rápó & Tonk, 2017). The spontaneity of the uptake process is determined by the alteration of entropy and energy values that are caused by corresponding variations in temperature. In other words, when there is an increase in Gibb's free energy with successive temperature variation and the spontaneity of the adsorption process is enhanced, it implies an endothermic initiated process that allows the dye particles to move swiftly to dynamic spots on the nanomaterials as temperature increases due to activation (Panda et al., 2021).

Bozbas research group observed the impact of temperature on adsorption capability for malachite green oxalate (MGO) and MB in water solution when green nanoparticle adsorbent was prepared using menengic coffee biowaste. The temperature influence was determined via the Van't Hoff equation for expressing the relationship among standard Gibb's free energy (ΔG°) and equilibrium constant, which negative values were obtained for adsorption entropy (ΔS°) and ΔG° toward the adsorption of MGO and MB, indicating that the removal mechanism for both dyes occurs spontaneously and exothermically (Bozbas, Aras, Karabulut, & Kayan, 2021).

In another research by Einafshar and colleagues, two nanomaterials with multifunctional ability were prepared utilizing graphene sheet, TiO_2 and $\text{Ag}/\text{Al}_2\text{O}_3$ that were incorporated with β -cyclodextrin-GO to form $\text{Ag}/\text{Al}_2\text{O}_3/\text{TiO}_2@ \beta$ -cyclodextrin-GO (AATG) and $\text{TiO}_2@ \beta$ -cyclodextrin-GO (TG) that were comparatively examined toward the sequestration of MB dye. In the experiment, the effect of temperature toward adsorption efficacy was evaluated at temperatures from an array of 298–328K. From the experiment, MB was uptaken efficiently at elevated temperatures, implying that the adsorption mechanism relied on temperature that makes the reaction endothermic. With a temperature rise, the dye particles acquire higher kinetic energy to bond with vacant binding spots on the exterior part of the nanomaterials enhancing the removal efficiency above 99% (Einafshar et al., 2023).

Another research by Mahmoud investigated the impact of temperature on the elimination efficacy of direct yellow 50 dye from textile effluent using nanobentonite adopting varying temperatures (K) (293, 303, 313, 323, 333, and 343) and obtained corresponding removal efficiency for each temperature. It was observed that 303K was the most efficient

TABLE 19.3 Effects of temperature on elimination efficacy of dyes in water using nanomaterials.

Nanomaterial	Dye adsorbed	Temperature (K)	Efficiency (%)	Reaction type	Quantity in equilibrium (q_e , mg/g)	References
Poly(acrylonitrile-styrene) Multiwalled carbon Nanotubes	Reactive red 35	298–328	63.33–9.07 67.55–97.61	Exothermic Endothermic	– –	Abualnaja, Alprol, Abu-Saied, Ashour, & Mansour (2021)
Fe ₃ O ₄ loaded on ash from plant leaves	Malachite green	298–333	73.22–80.65	Endothermic	Increased 169.21–244.67	Agarwal et al. (2016)
CG-Zn (OH) ₂	Coomassie violet	299–333	47.86–89.94	Endothermic	Increased 40.26–52.58	Parimelazhagan et al. (2023)
OA-JS@ZnFe ₂ O ₄	Congo red Methylene blue	283–323	93.81–98.23 81.97–93.54	Endothermic	–	El Messaoudi et al. (2022)
Mycosynthesized Fe	Congo red Direct blue-1	298–308	98.5 82.8	Endothermic Endothermic	Increased 46.7–49.2 Increased 27.2–41.4	Singh et al. (2022)
Fe ₃ O ₄ @AC	Methylene blue Brilliant 27 green	283–323	81–93.5 76.94–92.1	Endothermic	Increased	Joshi et al. (2019)
Red alga A.- ZnO	Direct blue 106	298–318	–	Endothermic	370.37	Eleryan et al. (2023)
P@SiO ₂	Methylene blue	298–353	–	Exothermic	Decreased 76.92	Nayl et al. (2022)
Modified zeolite A	Congo red	297–309	–	Exothermic	Decreased 21.11	Khalaf, Al-Sudani, AbdulRazak, Aldahri, & Rohani (2021)
Ze/AC Ze/L/AC	Methylene blue	294–313	90 92	Endothermic	Increased	Elmekawy, Quach, & Abdel-Fattah (2023)

temperature with a removal efficiency of 94.6%, but further increase resulted in a decrease in the efficacy that proposed the adsorption mechanism to be exothermic (Mahmoud, 2022). Other studies on the impact of temperature on the adsorption capability of dyes using nanomaterials are presented in Table 19.3.

19.2.4 Regenerability and recyclability

From the sustainable eco-economical angle of adsorption operation and other dye treatment techniques, one of the key factors in defining an adsorbent's effectiveness is regeneration (Bayode, Olisah, Emmanuel, Adesina, & Koko, 2023). The regeneration (desorption of adsorbed pollutant) operation allows for the restoration of architectural integrity and adsorption capacity of expended adsorbent, and the restoration prospects determine the adsorbent reusability for cost-effective real-life wastewater treatment application (Moghadas, Motamedi, Nasiri, Naghavi, & Sabokdast, 2020; Vakili et al., 2019; Wang & You, 2021). As seen from the literature, different methods in a combo or singly are employed for regeneration operation; however, the chemical method is the most come one owing to cost-effectiveness, simplicity, and rapid process benefits compared to other methods that are difficult to scale up, cause adsorbent weight loss (thermal

and ultrasonic treatments), and release of hazardous gases (thermal treatment) upon heating or even birth explosion due to uncontrolled heating (microwave irradiation) (Abdullah et al., 2019). However, some of the inorganic solvents employed with the chemical method tend to destroy the surface profile of adsorbents just like heat also does in the thermal method nanomaterials (Abdullah et al., 2019). Nonetheless, this demerit, alongside the issue of eluent/regenerate disposal, has made researchers concentrate more efforts on using water and other less toxic organic solvents as eluent/regenerate in combination with magnetic separation at time (Kataria & Garg, 2019; Zhou, Lu, Zhou, & Liu, 2019). Table 19.4 gives a summary of the adsorption performance of various expended nanomaterials and their derivative biocomposite for dye pollutants removal after regeneration up to nth rounds.

19.2.5 Mechanism of dye adsorption onto nanocomposite

Adsorption is a surface phenomenon that occurs when a substance from its liquid or gaseous surroundings concentrates or accumulates at a solid interface or surface (adsorbent) to the bulk phase (Pourhakkak, Taghizadeh, Taghizadeh, Ghaedi, & Haghdoost, 2021). By and large, there are two categories of adsorption: physisorption (when there is a reversible, weak van der Waals force-based physical interaction between the adsorbate and the adsorbent) (Borodin et al., 2020; Ma et al., 2022; Zhou et al., 2019) and chemisorption (a stronger chemical reactions that arise when the nanocomposite has active groups or spots that may form covalent connections with the adsorbate) (Agboola & Benson, 2021; Borodin et al., 2020; Sanghi & Verma, 2013; Umeh et al., 2023). However, in the chemistry of dye removal, the most essential task is selecting the best propitious nanosorbent materials, specifically those with the greatest combination of strong selectivity, affordability, superb adsorption efficiency, and quick kinetics, and to conquer these challenges, a solid understanding of the interplay between adsorbates like dye and nanocomposite is a prerequisite (Bayode et al., 2020). Additionally, comprehending the adsorption mechanism will provide valuable insight into developing improved nanobiocomposite and exceptional desorption strategies for recovering the decontaminated dye and making spent nanobiocomposite recyclable (Crini et al., 2019). Generally, the adsorption operation encompasses the materialization of a monolayer accompanied by the development of multiple layers, with monolayer formation taking place quickly and intraparticle diffusion progressing at a slower rate, while the mechanism of adsorption of dye on nanocomposite is typically through electrostatic interaction, weak van der Waals interaction, π - π interaction, complexation, ion exchange, H-bonding, or hydrophobic interaction (Bayode et al., 2023; Umeh et al., 2023). Nonetheless, the adsorption operation is driven by one or a combo of two or more of the aforementioned interactions (Bayode et al., 2023). Also, many parametric factors govern the mechanism, and this includes the temperature and pH of the solution, nanocomposite architectural functional groups and the textural profile of the nanocomposite, dye dissolution rate, and the molecular structure of the target dye (Akpomie & Conradie, 2020; Ambaye, Vaccari, van Hullebusch, Amrane, & Rtimi, 2021; Bayode et al., 2020; Rojas & Horcajada, 2020). It is also interesting to say that in some situations, kinetics and isotherm models, thermodynamic parameters, and other models can be used to unravel the mysteries of nanocomposite-dye interactions and determine the most likely adsorption mechanism route (Iwuzor et al., 2022). Although it is difficult to identify the precise interactions at play, various scientists find the fourier transformed infrared spectroscopy (FTIR) spectroscopy to be a handy technique for exploring the interplay between nanocomposite-dye during adsorption operation (Akpomie & Conradie, 2020; Hosseinzadeh & Mohammadi, 2015; Sharma, Mangla, Choudhry, Sajid, & Ali Chaudhry, 2022; Siddiqui & Chaudhry, 2018). Interestingly, since surface functional groups have a significant impact on the adsorption mechanism, the process might be arbitrarily predicted based on surface charge and FTIR (Akpomie & Conradie, 2020; Siddiqui & Chaudhry, 2018).

For example, Siddiqui and Chaudhry (2018) opined that the existence of a significant number of architectural surface functional sites in the biofabricated nanocomposite $\text{MnFe}_2\text{O}_4/\text{BC}$ was the reason for its greater MB adsorption capacity when compared to the other adsorbents. The interaction between MB and $\text{MnFe}_2\text{O}_4/\text{BC}$ caused some shifting in the positions of FTIR peaks following MB adsorption, the authors emphasized. These shiftings corresponded to $-\text{C}-\text{O}-$ stretching, symmetric, and asymmetric stretching of $-\text{NH}_2$ and $-\text{COO}$ groups (Siddiqui et al., 2018). The bands at 1709 and 1660 cm^{-1} entirely vanished, which might be related to MB binding to the amide and ketonic groups, correspondingly. The reported change in the stretching frequency of $-\text{OH}$ was also attributed to H-bonding between the MB and $\text{MnFe}_2\text{O}_4/\text{BC}-\text{OH}$ following MB adsorption. However, there was no discernible shift in the location of the FTIR peaks that corresponded to the $\text{C}-\text{H}$ stretching frequency (Siddiqui & Chaudhry, 2018). This demonstrated a specific electrostatic interaction and H-bonding between the cationic MB and $\text{MnFe}_2\text{O}_4/\text{BC}-\text{OH}$ active sites.

In another study, PVP@CNTs- Cu_2O nanocomposite and malachite green interaction were subjected to FTIR and xray photoelectron spectroscopy (XPS) studies, which demonstrated the involvement of surface carbonyl and OH functional groups in the adsorption operation (Li, Dong, & Zhao, 2021). The bands for $\text{C}=\text{C}/\text{C}-\text{C}$ were seen to shift to

TABLE 19.4 Summary of the adsorption performance of various expended nanomaterials and their derivative biocomposite for dye pollutant removal.

Nanobiocomposite adsorbent	Dye adsorbed	Regenerant	% Adsorbed at first cycle	No. of cycles (n)	% adsorbed at nth cycles	References
Cress seed mucilage@Fe ₃ O ₄	MB	0.5 M HCl	84	3	67	Allafchian et al. (2019)
<i>Ocimum sanctum</i> @Fe ₃ O ₄	MB	0.5 M HCl + magnetic shaken	94.23	5	69.74	Sharma et al. (2022)
Fe ₃ O ₄ @chitosan@GO@PEI	CR	–	>85	5	>80	Li et al. (2021)
Fe ₃ O ₄ @chitosan@GO@PEI	Amaranth	–	>85	5	>80	Li et al. (2021)
Organo-bentonite/Co ₃ O ₄	MG	Distilled H ₂ O	99.80	5	77.40	Abdel Salam et al. (2020)
Fe ₃ O ₄ @sugar-beet pulp CNCs/starch-g-(AMPS-co-AA) hydrogel	MB	Diluted aqueous HCl (pH 4)	~ 100	5	95.6	Moharrami and Motamedi (2020))
Fe ₃ O ₄ @ <i>Musa acuminata</i> peel	Bromophenol blue	0.5 M NaOH	80.3	4	57.4	Akpomie and Conradie (2020)
ZnO@BC sugarcane bagasse	MO	Distilled H ₂ O	90.8	6	86.2	Gonçalves et al. (2020)
Carboxymethyl cellulose/alginate/polyvinyl alcohol/rice husk	Direct red-31	Distilled H ₂ O (pH 12)	>85	4	~ 40	Bhatti et al. (2020)
ZIF-8@Fe/Ni	MG	EtOH	99.00	5	73.00	Zhang, Jin, Owens, & Chen (2021)
Fe ₃ O ₄ /PT/GO	MG	50% HCl	94.50	5	85.90	Gao et al. (2019)
Fe ₃ O ₄ @ <i>Lonicera japonica</i> flower powder	MB	Magnet + pure acetonitrile	~ 100	7	~ 95	Lingamdinne, Karri, Khan, Chang, & Koduru (2021)
Sugarcane bagasse CNF@Fe@FeS	CR	0.5 N NaOH and H ₂ O	71.5	5	70.6	Sankararamkrishnan, Singh, & Srivastava (2020)
Chitosan-ZIF-2	MG	EtOH	~ 100	5	–	Amin, Shojaei, & Hamzehlouyan (2022)
ZnO@ <i>Ananas comosus</i> waste	Celestine blue	0.2 M HCl	51.6	4	40.8	Akpomie and Conradie (2020)
Fe@ <i>Eleocharis dulcis</i> BC	CR	HCl (5% v/v) and distilled H ₂ O	84.4	5	79.9	Imran, Javed, Areej, & Haider (2022)
nZV Cu@ <i>Eleocharis dulcis</i> BC	CR	HCl (5% v/v) and distilled H ₂ O	91.8	5	87.9	Imran, Javed, Areej, & Haider (2022)
g-C ₃ N ₄ /Fe ₃ O ₄ /ZIF-8	MG	EtOH	100	5	100	
Ni/Al@rice husk BC	CR	–	78	5	58	Siregar, Palapa, Wijaya, Fitri, & Lesbani (2021)
Fe ₃ O ₄ @sugar-beet pulp CNCs/starch-g-(AMPS-co-AA) hydrogel	CV	Diluted aqueous HCl (pH 4)	~ 100	5	96.2	Moharrami and Motamedi (2020)

(Continued)

TABLE 19.4 (Continued)

Nanobiocomposite adsorbent	Dye adsorbed	Regenerant	% Adsorbed at first cycle	No. of cycles (n)	% adsorbed at nth cycles	References
Zn/HAP/MgFe ₂ O ₄	MG	Magnet	100	7	95	Das & Dhar (2020)
Montmorillonite clay/PANI/Fe ₃ O ₄	MB	0.5 M HCl	100	5	100	Mu, Tang, Zhang, & Wang (2016)
Waste scrap tires AC/Fe/Ce	RhB	5 mL EtOH	95	7	89	Tuzen, Sari, & Saleh (2018)
rGO/TiO ₂ /acrylonitrile-co-maleic	MG	Acetonitrile-formic acid	>95	20	>80	Du et al. (2020)
Sugarcane bagasse CNF@Fe@FeS	MB	0.5 N NaOH and H ₂ O	98.9	5	97.6	Sankaramakrishnan, Singh, & Srivastava (2020)
PDA/Fe ₃ O ₄ /GO/CA-β-CD	MG	0.5 M HCl	~94	8	~83	Yan, Li, Yan, Fang, & Liu (2022)
PDA/Fe ₃ O ₄ /GO/CA-β-CD	CV	0.5 M HCl	~92	8	~87	Yan, Li, Yan, Fang, & Liu (2022)
PDA/Fe ₃ O ₄ /GO/CA-β-CD	MB	0.5 M HCl	~96	8	~88	Yan, Li, Yan, Fang, & Liu (2022)
Ag@tea leaf BC	RhB	0.1 M EDTA	80.90	5	67.48	Shaikh et al. (2022)
Ag@tea leaf BC	RhB	0.1 M HNO ₃	82.23	5	65.49	Shaikh et al. (2022)
Ag@tea leaf BC	CR	0.1 M NaOH	86.22	5	64.35	Shaikh et al. (2022)
Ag@tea leaf BC	CR	0.1 M EDTA	84.39	5	61.44	Shaikh et al. (2022)
Chitin@MgO	MO	0.5 M NaOH	95.4	5	90.7	Majdoubi et al. (2023)
Pearl millet stem@MWCNT	SO	Acetone + hot H ₂ O	98.81	10	94.45	Yadav, Bagotia, Yadav, Sharma, & Kumar (2022)
Pearl millet stem@CNT	MB	Acetone + hot H ₂ O	98.11	10	93.45	Yadav, Bagotia, Yadav, Sharma, & Kumar (2022)

AC, Activated carbon; starch-g-(AMPS-co-AA), 2-acryl amido-2-methyl propane sulfonate and acrylic acid; BC, biochar; CA, citric acid; CD, cyclodextrin; CNF, cellulose nanofiber; CNT, carbon nanotubes; CV, crystal violet; CR, Congo red; EDTA, Ethylenediaminetetraacetic acid; GO, graphene oxide; HAP, hydroxyapatite; HCl, hydrochloric acid; MB, methylene blue; MWCNT, multi walled carbon nanotube, nZV, nano zero-valent; PANI, polyaniline; PDA/MGO/CA-CD = PDA, polydopamine; PEI, polyethyleneimine; PT, persimmon tannins; RhB, rhodamine B; SO, safranin O; ZIF, zeolitic imidazolate framework.

binding energies of greater value (284.82 eV) following malachite green adsorption, indicating a π - π interaction between malachite green's benzene rings and the CNTs skeleton in nanobiocomposite (Lin & Chang, 2015). The possible H-bonding between carbonyl functional groups and malachite green is shown by the change of the C = O bond from 287.63 to 288.37 eV. O1s spectra corroborated these findings. Two Cu^+ peaks migrated to higher levels in a ΔBE range of 0.25–0.28 after malachite green adsorption, indicating the production of metal oxide nanocomposite-malachite green complexes involving Cu–O bonds and organic pollutants. Oxygen atoms in these complexes served as malachite green's reaction spots, which decreased the electron cloud density around Cu atoms and raised binding energy (Umeh et al., 2023). In addition, nanocomposites modified with groups such as OH and $-\text{NH}_2$ groups or modified surfaces show chemisorption (Ali, Al-Othman, & Alwarthan, 2016; Umeh et al., 2023).

In another study, Moharrami's research group (Moharrami & Motamedi, 2020) used Langmuir isotherm to propose a physisorption mechanism for dye uptake onto nanocomposite. Conversely, El-Zahhar, Awwad, and El-Katori (2014) observed that the comparatively large $-ve \Delta S^\circ$ values indicate a drop in dye concentration at the nanobiocomposite–solution interface, which in turn indicates an upsurge in dye concentration on the solid nanobiocomposite phase. It was then mechanistically further accentuated that the result is a typical byproduct of the chemisorption process, indicating that the sorption of bromophenol blue dye on the polymer clay nanocomposite adsorbent was chemical in origin (El-Zahhar et al., 2014) at pH levels >5.77 , the carboxylic groups on the hybrid nanocomposite deprotonate, forming positively charged COO^- ligands. The ligands then bind to $+vely$ charged molecules of MG, indicating that ion exchange is the mechanism behind the adsorption of MG by the nanocomposite (Abu Elella et al., 2021; Kumar, Dhami, Rehani, & Singh, 2022).

As for the role of pH in adsorption mechanism elucidation when the pH of the solution rises over pH_{zc} , the nanocomposite surface potential becomes extremely $-ve$, which amplifies the electrostatic effect, while the nanocomposite surface charge becomes positive when the pH drops below pH_{zc} , making it difficult to adsorb $+vely$ charged dye molecules (Akpomie et al., 2023). For instance, in a particular study (Allafchian, Mousavi, & Hosseini, 2019), the positively charged chitosan modified (CSM)-based nanocomposite particles related to $-ve \text{COOH}$ groups show a decreased rate of adsorption at pH between 2 and 4, while the dye adsorption is seen to peak at pH levels between 4 and 9. The reason for that is explained by the abundance of H^+ species at pH values lower than 4, which would effectively compete with the adsorption of positively charged MB dye molecules on the nanobiocomposite surface. On the other hand, the dominating electrostatic interaction between dye molecules and nanobiocomposite particles would speed up the adsorption process and improve the adsorption aptitude at $\text{pH} >4$, when the available $-vely$ charged nanobiocomposite interface is enhanced. As a result, it was deduced that the adsorbent would adsorb more $+vely$ charged MB dye molecules because of the presence of $-ve$ functional groups on its interface at pH values greater than pH_{zpc} (Allafchian et al., 2019). The explained adsorption mechanism was consistent with the one expounded by other researchers from similar studies (Abdel Salam, Abukhadra, & Adlii, 2020; Akpomie & Conradie, 2020; Hosseinzadeh & Mohammadi, 2015; Moharrami & Motamedi, 2020; Sharma et al., 2022). Finally, thermodynamics also plays a significant part in comprehending the adsorption mechanism by characterizing the kinds of interactions depending on the size of ΔG value. The ΔG values indicate chemisorption when they are $>40 \text{ kJ/mol}$ and indicate physisorption when they are $<40 \text{ kJ/mol}$ (Umeh et al., 2023).

19.2.6 Outlook

Need for standardized testing protocols to accurately compare the performance of different nanomaterials and biocomposites under varying conditions, aiding in the establishment of robust guidelines for practical implementation. While these materials offer promising solutions, it is imperative to assess their long-term impact on the ecosystem to ensure that the benefits of pollution mitigation are not overshadowed by unintended consequences.

19.3 Conclusion

In conclusion, the utilization of nanomaterials and their derivative biocomposites for dye adsorption represents a promising avenue for addressing environmental pollution. The unique properties of nanomaterials, combined with the biocompatible nature of composites, offer a sustainable and efficient solution for dye removal from various sources. The journey in this field has witnessed significant advancements, from understanding the fundamental principles of adsorption to the development of sophisticated nanocomposites. However, challenges remain, such as ensuring long-term stability, addressing potential toxicity concerns, and optimizing the scalability of production. As researchers continue to explore new frontiers, the interdisciplinary nature of this field will likely lead to exciting collaborations among

materials scientists, chemists, environmental engineers, and biotechnologists. The continuous pursuit of knowledge in nanomaterials and their biocomposites for dye adsorption holds the promise of cleaner water systems and a healthier environment for future generations.

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