

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/380389332>

Greenly synthesised novel microporous ZnO/GO for adsorption of methyl orange and malachite green

Article in International Journal of Environmental Analytical Chemistry · May 2024

DOI: 10.1080/03067319.2024.2346198

CITATIONS

5

READS

47

3 authors, including:



Anuj Sharma

Guru Jambheshwar University of Science & Technology

12 PUBLICATIONS 108 CITATIONS

SEE PROFILE



Dr Mona Sharma

Central University of Haryana

62 PUBLICATIONS 978 CITATIONS

SEE PROFILE



Greenly synthesised novel microporous ZnO/GO for adsorption of methyl orange and malachite green

Anuj Sharma ^a, Sharma Mona ^b and Praveen Sharma ^a

^aDepartment of Environmental Science and Engineering, Guru Jambheshwar University of Science & Technology, Hisar, India; ^bDepartment of Environmental Studies, School of Interdisciplinary and Applied Sciences, Central University of Haryana, Mahendergarh, India

ABSTRACT

This study presents a greenly synthesised novel nanocomposite composed of Zinc Oxide (ZnO) and Graphene Oxide (GO) for the adsorption of dyes from aqueous solutions. The nanocomposite was synthesised using algae thus representing a green and eco-friendly method, ensuring minimal environmental impact. The physical characterisation of zinc oxide and graphene oxide nanocomposite (ZnO/GO) was done using X-ray diffraction (XRD), Brunauer-Emmett-Teller (BET), Fourier transform infrared spectroscopy (FT-IR), Zeta Potential, and Particle Size Analyzer (PSA). The pore diameter of the ZnO/GO was 1.801 nm, and its total surface area was 54.062 m²/g. The nanocomposite's adsorption performance was systematically investigated through batch adsorption experiments. With an adsorption efficiency of 296.73 mg/g and 259.74 mg/g for MO and MG, the Langmuir isotherm model outperforms the Freundlich isotherm model in terms of adsorption. The pseudo-second-order model having an R² value of 0.99 for both MO and MG, offers a more convincing explanation of adsorption than the pseudo-first-order model. The results demonstrated rapid adsorption kinetics and high adsorption capacities for both cationic and anionic dyes. Additionally, the ZnO/GO has a high reusability without significant degradation. The sustainable nature of the nanocomposite was emphasised by its efficient dye removal, reusability, and minimal secondary waste generation.

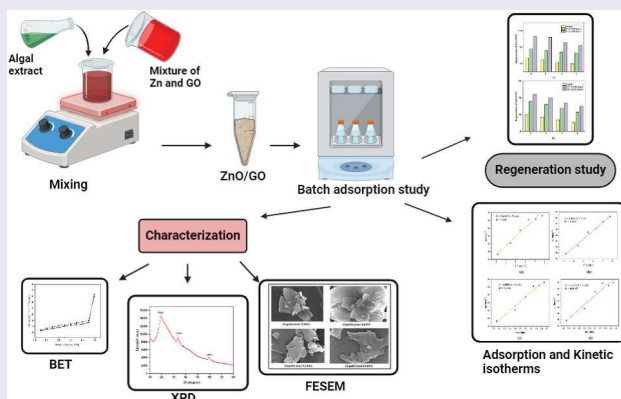
ARTICLE HISTORY

Received 22 March 2024

Accepted 17 April 2024

KEYWORDS

Wastewater; zinc oxide; graphene oxide; nanocomposite; remediation; dye



CONTACT Praveen Sharma  ps.enbt@gmail.com

 Supplemental data for this article can be accessed online at <https://doi.org/10.1080/03067319.2024.2346198>.

© 2024 Informa UK Limited, trading as Taylor & Francis Group

1. Introduction

The usage of synthetic dyes and the fast growth of industry have resulted in a startling rise in environmental contamination. Both cationic and anionic dye contaminants are well known for their harmful impacts on water bodies, which puts aquatic ecosystems and public health at significant risk. Physical and chemical methods utilised for wastewater treatment have played a significant role in mitigating water pollution; however, they are associated with numerous drawbacks in terms of their ecological impacts. Here are some of the main shortcomings of these techniques: production of sludge, chemical leakage, production of hazardous byproducts, energy intensity, disturbance of habitat, non-selective removal, acidification and pH imbalance in the environment, environmental persistence, and interference with natural processes [1,2]. However, the emergence of bionanotechnology has brought forth a new paradigm with several distinct advantages over traditional techniques. The following are some main advantages of treating wastewater with bionanotechnology: improved selectivity and efficiency, less need for chemicals, less energy use, increased adaptability and versatility, less sludge production, the potential of resource recovery, environmental compatibility and biodegradability, synergistic effects, and advancements in monitoring and detection [3–10]. In the field of nanomaterial-based adsorbents, the need for efficient and sustainable dye remediation techniques has spurred significant research. Of them, nanocomposites have shown great promise because of their adjustable characteristics and increased adsorption capacity. In this regard, the ZnO/GO synthesis offers a novel strategy for dealing with the removal of dyes from water. Zinc oxide (ZnO) and Graphene oxide (GO) are two different nanomaterials with unique properties. On the one hand, GO has a two-dimensional structure and a large number of functional groups, while ZnO is a semiconductor material with a high surface area and photocatalytic activity. These substances working together as a nanocomposite could improve their adsorption qualities in a complementary way. Importantly, the green synthesis approach adopted in this study minimises the use of dangerous chemicals and energy-intensive procedures, ensuring environmentally friendly production [5,11–13].

In this study, the algal extract is used as a reducing agent. Various proteins, essential fatty acids, vitamins, pigments, and polysaccharides were present in algal extracts or biomolecules that act as capping and reducing agents during the synthesis of nanoparticles. This technique offers numerous advantages over conventional methods that use chemicals, including the absence of toxic by-products, mild reaction conditions, and biocompatibility. One of the main advantages of utilising algae in synthesising nanoparticles is their renewable nature and it can be easily cultivated. The advantages of this method make it a sustainable alternative to the chemical methods of synthesising nanoparticles [14–19]. The application of greenly produced ZnO/GO for the adsorption of dyes holds several significant advantages. Firstly, the environmentally friendly synthesis method reduces the negative effects of nanomaterial production on the environment, in line with the principles of sustainable development. Secondly, the nanocomposite's enhanced adsorption capability and adjustable characteristics offer a versatile solution for the removal of various kinds of dye contaminants. Finally, the

understanding of the adsorption mechanisms advances our understanding of the behaviour of nanocomposite materials in water treatment applications [1,20,21]. The development of effective and sustainable water treatment technologies is imperative due to the growing global concern over water scarcity and contamination. The ZnO/GO offers a promising solution to dye pollution because of its remarkable adsorption capabilities. By adsorbing both cationic and anionic dyes, it offers a comprehensive solution to water bodies contaminated with a varied variety of dye contaminants. ZnO/GO holds great potential in real-world scenarios of water treatment. It is a viable option for large-scale implementation due to its scalability, ease of synthesis, and compatibility with existing water treatment processes. Furthermore, the economic feasibility and sustainability of the nanocomposite are enhanced by the possibility of regenerating the nanocomposite for multiple adsorption cycles through desorption and reuse strategies. The pressing issue of dye pollution in water bodies must be addressed by developing innovative, sustainable, and efficient adsorbents [2,5,13,22]. The environmentally friendly synthesis of ZnO/GO presents a significant step towards sustainable development. A versatile platform for dye removal is provided by the unique combination of GO's adsorption capabilities and ZnO's photocatalytic capabilities.

This work contributes to the understanding of the behaviour of nanocomposite in wastewater treatment applications and establishes the groundwork for future investigation and advancement in the field of sustainable environmental remediation. The objectives of the current study were (i) synthesise ZnO/GO employing environmentally friendly reducing agents. (ii) Characterisation of ZnO/GO. (iii) Absorption of MO and MG on ZnO/GO. (iv) Assessing the adsorption using kinetics, thermodynamics, and the adsorption isotherm, thereby understanding the adsorption mechanism. (v) Desorption and regeneration studies.

2. Materials and methodology

2.1. Materials

In the current experimental study, chemicals bearing the mark of standardised methodology were used; no refining procedure was used for the chemicals. Zinc acetate ($\text{ZnC}_4\text{H}_6\text{O}_4$) (Mol. Wt. = 183.48 g/mol), Graphene Oxide (NbO_2) (Mol. Wt. = 124.91), algal extract, ethanol ($\text{C}_2\text{H}_5\text{OH}$), and all solutions generated using double distilled water. The pH of the dye solution was managed using 0.1 N HCl and 0.1 N NaOH.

2.2. ZnO/GO synthesis

2.2.1. Algal extract

3.5 g of algal biomass was added to 50 ml of distilled water with continuous stirring for 6 hours at 75°C. The solution was filtered out and stored for further processing to synthesise ZnO/GO.

2.2.2. Preparation of ZnO/GO

GO was synthesised using a modified Hummer's method [12]. 0.1 g of GO was mixed in 100 ml of distilled water followed by ultrasonication for 1 hour. 0.5 M solution of zinc acetate was prepared, then 100 ml of 0.5 M solution of zinc acetate was added to 100 ml of sonicated GO solution, then the mixture was stirred continuously for 2 hours at 60°C. Hereafter 100 ml of this mixture of 0.5 M zinc acetate and GO was mixed with 100 ml of algal extract followed by continuous stirring for 2 hours at 70°C and then allowed to cool. Once the ZnO/GO were synthesised the mixture was washed with ethanol and centrifuged five times at 6000 rpm. The now-obtained ZnO/GO was dried for 6 hours at 70°C in the oven, and then with the help of a motor and pestle, they were crushed. Thus ZnO/GO were synthesised, which were then prepared for use in the removal of MO and MG. Fig. S1. depicts the methodology used for synthesising ZnO/GO in flow chart form.

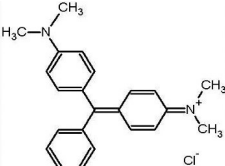
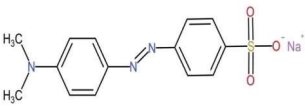
2.3. Dye solutions

The MG and MO were each separately dissolved in 1000 mL of double distilled water to create the stock solution. Concentrations from 10 mg/L to 80 mg/L were created using this solution. Peculiar attributes of MO and MG are depicted in Table 1 [23–25].

2.4. Characterization of ZnO/GO

These strategies were utilised for the characterisation of ZnO/GO. XRD was done utilising Benchtop Miniflex- II Rigaku at a λ of 1.54056 Å, scale of 2θ was 10 ° to 90 °. FTIR was done utilising the Perkin Elmer model, having KBr power for the collection of data, 4000–500 cm^{-1} is the spectrum range, and 0.5 cm^{-1} is the highest resolution. For zeta potential and PSA Malvern Nano-ZS90 was used. For FE-SEM analysis Model/Maker-7610F Plus/JEOL was utilised. UV-VIS spectroscopy was performed utilising the spectrophotometer Varian Cary-5000 respectively. BET was performed utilising Quantachrome Autosorb (iQ-MP),

Table 1. Peculiar features of malachite green and methyl orange.

	Malachite Green	Methyl Orange	References
Empirical Formula	$\text{C}_{23}\text{H}_{25}\text{ClN}_2$	$\text{C}_{14}\text{H}_{14}\text{N}_3\text{NaO}_3\text{S}$	
Molecular Weight (g/mol)	364.911	327.34	
λ_{max}	617	464	
pKa	10.3	3.46	
Applications	Agriculture, textile, and food sectors,	Textiles, printing, and pharmaceutical industries	[23,24]
Harmful effects	Carcinogenic, teratogenic, can cause reproductive abnormalities	Carcinogenic, teratogenic	[25]
Chemical structure			

2.5. Batch-adsorption experiments

By retaining the other parameters constant during the batch-adsorption analysis of ZnO/GO, a single parameter was varied. The pH was changed between 2 and 14 using 0.1 M NaOH and 0.1 M HCl, and the mixture was stirred continuously for 50 and 40 minutes for MG and MO correspondingly, at 200 rpm to examine the effects of pH. 0.02 g of ZnO/GO were mixed with 40 mg/l of MG and MO each for 50 and 40 minutes respectively. The absorbance was measured at a wavelength of 464 nm, and 617 nm for MO and MG respectively utilising UV-VIS spectrophotometry, to determine the effectiveness of removal. For assessing the impacts of ZnO/GO concentration, it was changed from 0.01 g to 0.05 g and all other variables remained constant followed by UV-VIS spectrophotometric analysis at respective wavelengths. To assess the effects of temperature, the temperature ranges between 20°C and 120°C. 0.02 g of ZnO/GO was used and at 200 rpm the solution was stirred. Utilising UV-VIS analysis, the remediation's efficiency was calculated. Further investigation was conducted using a thermodynamic study. To assess the effects of time, 0.2 g of ZnO/GO was mixed in a solution (500 mg/l) of MO and MG each, then swirled at 200 rpm for 40 and 50 minutes correspondingly. The remediation efficiency of ZnO/GO was determined utilising UV-VIS spectrophotometric analysis after withdrawing 50 ml of sample for each at intervals of 10 minutes. For further investigating the kinetic study, pseudo first and second, Bangham, IPDM, and Elovich were used. To test the influence of dye concentration, it varied from 10 mg/L to 80 mg/L while maintaining constant other parameters. The adsorption equilibrium was investigated using the Langmuir, Temkin, Freundlich, Dubinin-Radushkevich, and Redlich Peterson adsorption isotherms. Equations (S1) and (S2), respectively, were used to find the maximum capacity of adsorption and removal efficiency (%).

2.6. Regeneration study of ZnO/GO

A crucial feature that ensures the reusability of a nanomaterial is its regeneration capacity. Important information about the adsorption mechanism and nanomaterial reusability can be found in desorption research. For MO, pure ethanol, and ethanol having NaOH in 1 and 6% (v/v) were used as eluting agents for investigating the reusability of ZnO/GO, whereas for MG, 0.1 M HCl, 10% acetic acid, and pure acetic acid were used. The equation (S3) was used for calculating desorption (%).

3. Result and discussion

3.1. Physical characterization of ZnO/GO

3.1.1. XRD

The X-ray diffraction analysis of the ZnO/GO is depicted in Figure 1(a) and different parameters are described in Table 2, which shows the characteristic peak of zinc oxide with graphene oxide. The peaks were located at angles of 19.58°, 33.57°, and 59.40° on (2θ) and can be described with miller indices (hkl) values as (122), (134), and (452) crystalline planes of ZnO/GO on (2θ). The resulting peaks were verified using the standard

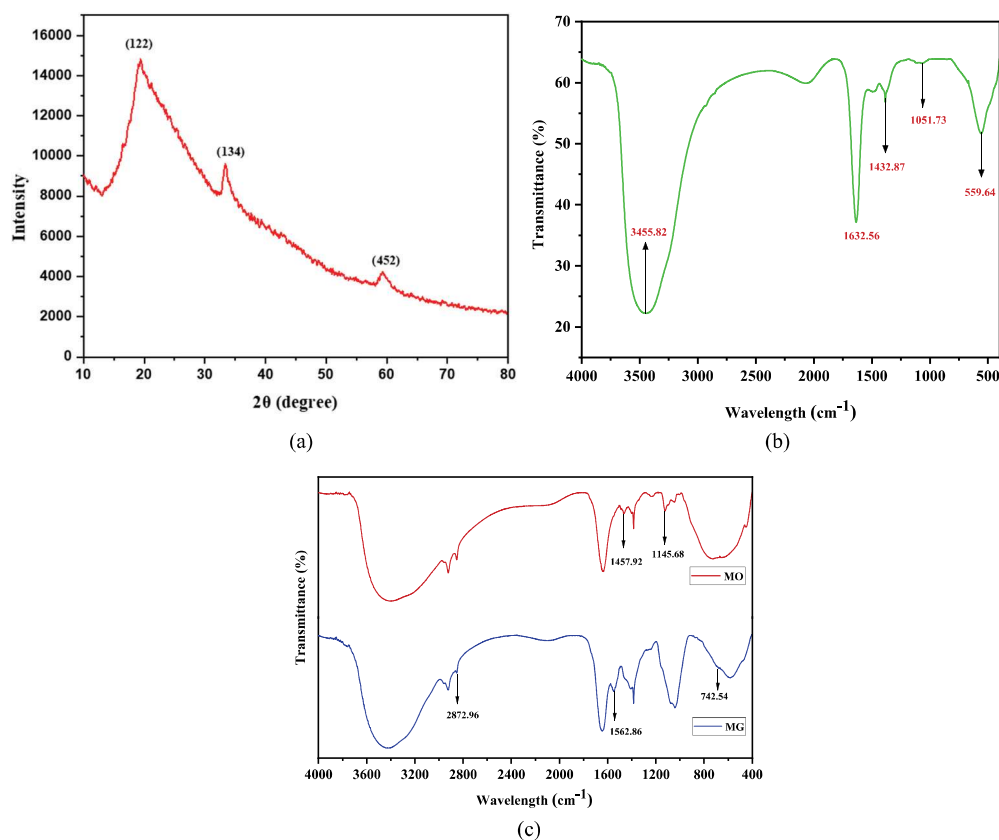


Figure 1. (a) XRD pattern of ZnO/GO. (b) FT-IR spectra of ZnO/GO. (c) FT-IR spectra of ZnO/GO loaded with MO and MG.

X-ray diffraction dataset JCPDS Card No: 4030757. A similar analysis was identified when zinc oxide/reduced graphene oxide nanocomposite was synthesised [26]. Equation (1) was used to get the crystalline index (%) for ZnO/GO. The crystalline index of ZnO/GO is 72.48%. Using the Scherrer Equation (2) the average size of ZnO/GO (17.982 nm) is calculated.

$$\text{Crystalline Index (\%)} = \frac{\text{Area of Crystalline peaks}}{\text{Area of all peaks}} \times 100 \quad (1)$$

$$d = K\lambda/\beta \cos \theta \quad (2)$$

3.1.2. FT-IR

Figure 1(b) depicts the FT-IR spectrum of ZnO/GO. FT-IR is employed to give details on the different functional groups and chemisorbed species that are present in the nanomaterial. The ZnO/GO nanocomposite spectrum observed peaks at 559.64 cm⁻¹ which depicts the stretching vibration of Zn-O deforms between ZnO atoms. The small wide peaks at 1051.73 cm⁻¹ describe the vibrational stretching of the C-O, and C-N bond which shows the presence of a tertiary alkoxy group which indicates the presence of phytochemical

Table 2. X-ray diffractometer data collection, lattice parameters, and peak shape of ZnO/GO.

Parameter	Value
Scan Speed	10.00 °/min
Step Width	0.02 °
CuK α Radiation	$\lambda = 1.5406 \text{ \AA}$
Scan Axis	$\theta/2\theta$
2θ Range	10°-80°
Lattice Parameter	$a = 8.57046 \text{ \AA}$, $b = 12.82023 \text{ \AA}$, $c = 16.97641 \text{ \AA}$ $\alpha = \gamma = 90^\circ$, $\beta = 101.30^\circ$
Peak Shape	Split pseudo-Voigt

compounds of algae which is beneficial for adsorption [2]. In the ZnO/GO spectrum, a small but sharp peak at 1432.87 cm^{-1} shows the presence of symmetric and asymmetric nitro compounds. In the ZnO/GO nanocomposite spectrum, sharp peaks were observed at 1632.56 cm^{-1} showing the vibrational stretching of C=O which describes the presence of symmetric carboxylate groups due to graphene oxide sheets [27]. The wide attributed transmittance in the ZnO/GO spectrum was observed at 3455.82 cm^{-1} which describes the stretching of the O-H group.

The FT-IR spectrum of ZnO/GO loaded with MG and MO dye is depicted in Figure 1(c). The additional peaks at 1145.68 cm^{-1} , indicate the existence of a strong acyl group and C–O stretching of anhydrides, while the second peak, located at 1457.92 cm^{-1} confirms the existence of asymmetric strong nitro groups, which indicates benzenoid. These groups confirm the adsorption of MO dye on the ZnO/GO surface [28]. Adsorption of MG on ZnO/GO has been confirmed by the appearance of recent peaks at 742.54 cm^{-1} , 1562.86 cm^{-1} , and 2872.96 cm^{-1} due to the presence of an asymmetric ring of hydrogen, benzene ring, and stretching vibration of C-H bond depicts the presence of aldehyde groups. A similar finding was observed during the adsorption of MG dyes onto nutraceutical industrial fenugreek seed spent (NIFGS) [29].

3.1.3. FE-SEM

The FESEM of ZnO/GO was depicted in Figure 2(a) which was used to examine the surface morphology to identify potential active sites for the adsorption of dyes. According to the FESEM study, ZnO/GO has wrinkles with a lateral dimension of 5–10 μm clearly showing the sheet-like structure of GO, indicating complete oxidation. The produced amorphous ZnO deposited uniformly on the surface of GO sheets, resulting in a flake-like shape. The crystal structure of the ZnO/GO was heterogeneous, containing rough areas. The EDAX of ZnO/GO is shown in Figure 2(b). The EDAX demonstrates the amount of zinc, carbon, and oxygen present in ZnO/GO. The EDAX of ZnO/GO displays that the amount of carbon weight was 41.34%, zinc weight was 38.03%, and oxygen weight was 20.63% present in ZnO/GO. The nanoparticles of zinc oxide on the surface of GO nanosheets were found to have a mean size of $46.78 \pm 2.14 \text{ nm}$. The FESEM measurements of ZnO particles were nearly identical to XRD measurements of crystallite sizes.

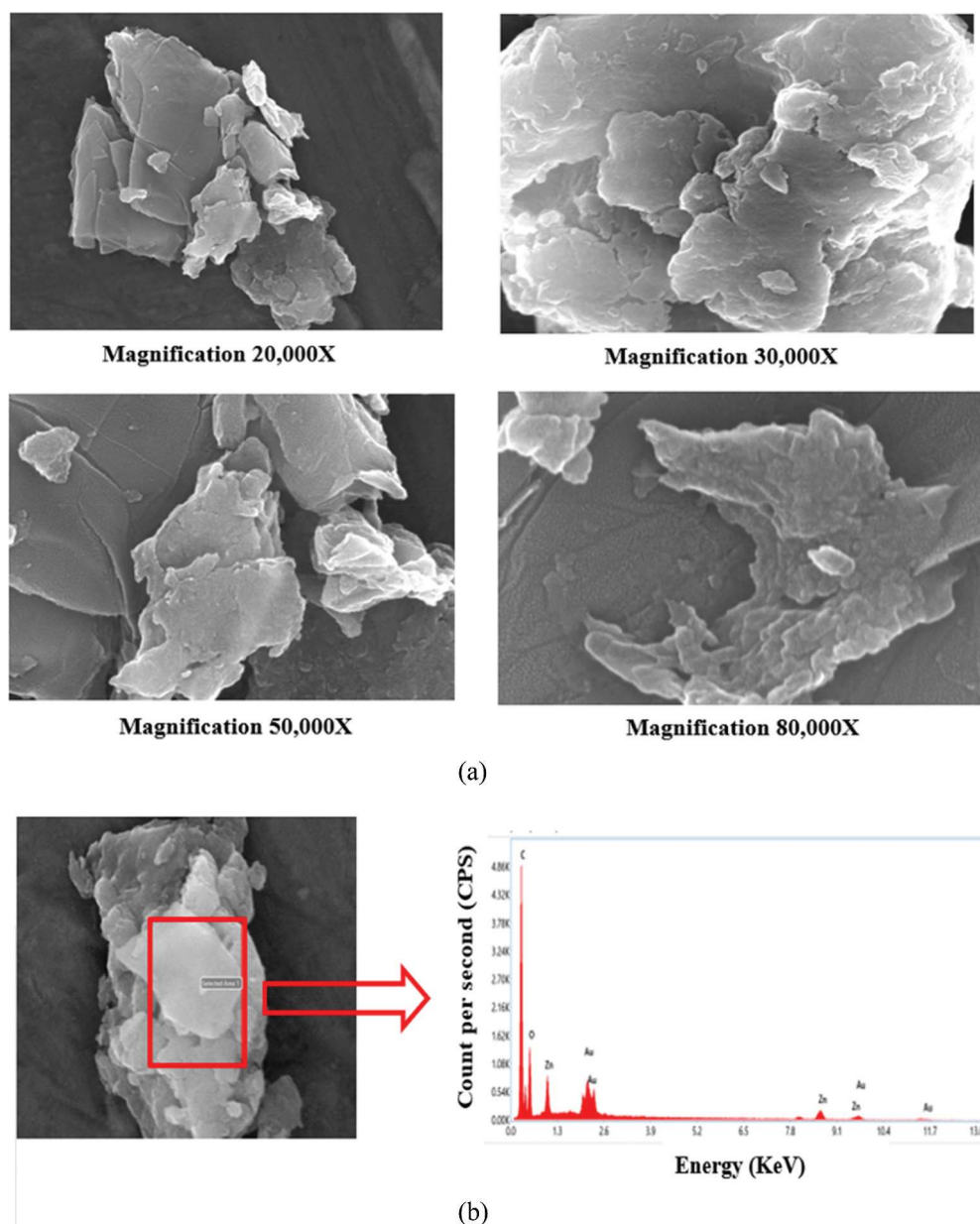


Figure 2. (a) FESEM image of ZnO/GO. (b) EDAX of ZnO/GO.

3.1.4. BET

Using BET analysis, the surface area and porosity of ZnO/GO were determined. BJH adsorption/desorption isotherms at liquid nitrogen (77.35 K) were used to calculate the surface area (Figure 3(a,b)). The ZnO/GO nanocomposite's specific surface area was 29.72 m²/g according to the BET study, but the total surface area of the pore, as determined by the BJH analysis, was 54.062 m²/g with a pore diameter of 1.801 nm, suggesting that the

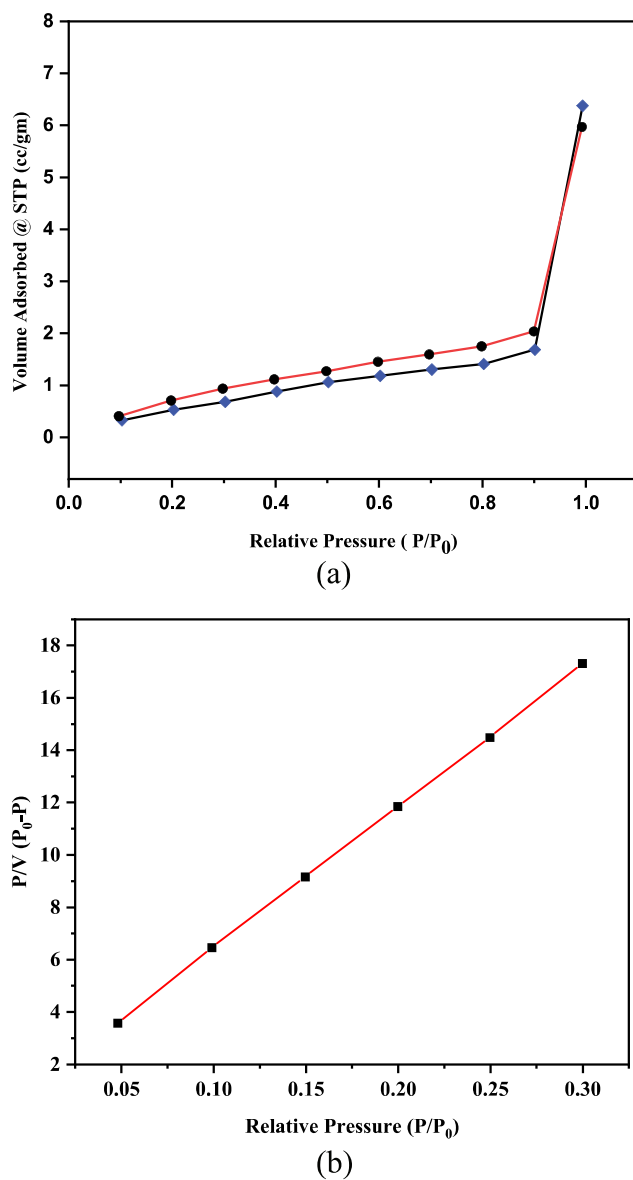
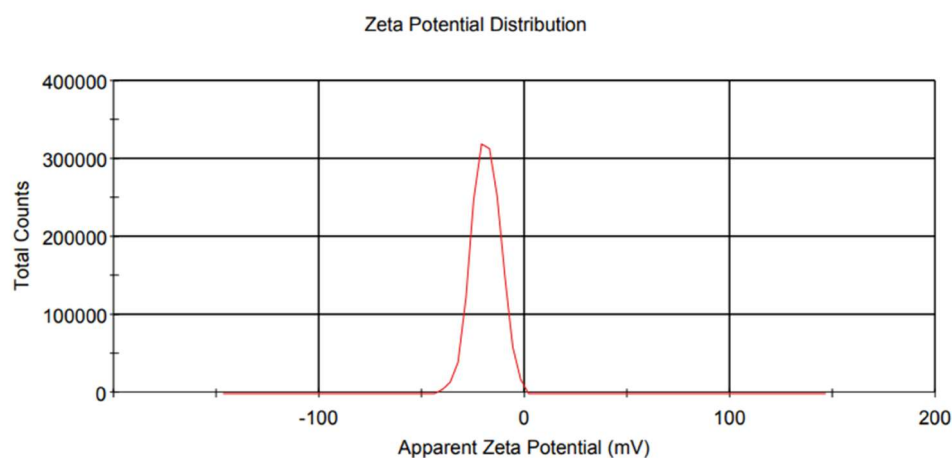


Figure 3. (a) Multi-point BET graph of ZnO/GO. (b) BJH adsorption/desorption isotherms graph of ZnO/GO.

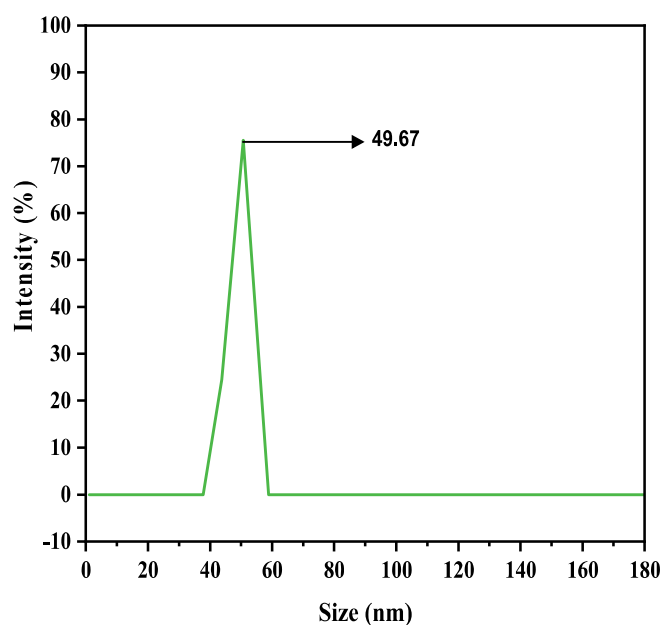
nanocomposite is microporous. ZnO/GO was discovered to have a cumulative pore volume of 0.146 cc/g.

3.1.5. Particle size analyzer and zeta potential

Zeta potential and PSA of ZnO/GO are depicted in Figure 4(a,b) which is used to determine the potential and size of the nanocomposite. According to the derived graphs, the zeta potential of ZnO/GO is -18.8 mV. The negative potential of nanocomposite facilitates high dispersity and excellent colloidal nature [18]. Biogenic synthesised reduced



(a)



(b)

Figure 4. (a) Zeta potential of ZnO/GO. (b) Particle size analyser (PSA) of ZnO/GO.

graphene oxide using an algal extract for the adsorption of pollutants showed a similar analysis [30]. The size of ZnO/GO applying PSA investigation was 49.67 nm which is similar to XRD Crystalline size.

3.1.6. AFM

Using AFM, the surface morphology of composite sheets was investigated. Figure 5(a,b) depicts the AFM images of the ZnO/GO. To properly measure the size of ZnO

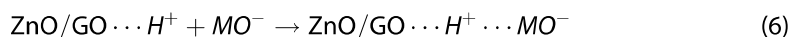
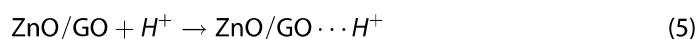
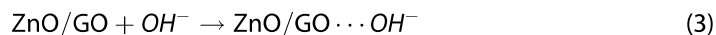
nanoparticles, AFM analysis was especially used for a portion of GO sheets adorned using well-dispersed inorganic elements. This revealed that the ZnO nanoparticles' average height was between 40 and 50 nm. The size of ZnO nanoparticles that were like the XRD and FESEM examination results that were discussed in the previous section. When zinc oxide/graphene oxide nanocomposite was synthesised for the degradation of fuchsin dye, similar results were obtained [31].

3.1.7. UV-VIS spectrum

The optical characteristics of ZnO/GO are investigated using UV-VIS spectroscopy, as illustrated in Fig. S2. The spectrum revealed a well-defined absorption peak associated with zinc oxide and graphene oxide. The peak of synthesised nanocomposite was detected as 356.24 cm^{-1} with absorbance at 1.42 which corresponds to $n\text{-}\pi^*$ transition of $\text{C}=\text{O}$. A similar peak was observed during the synthesis of ZnO NPs with graphene oxide [32].

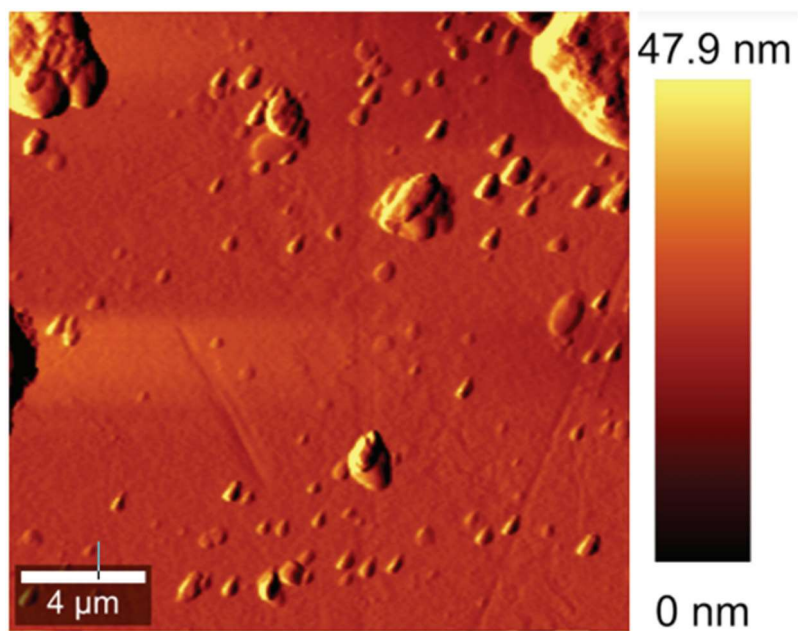
3.1.8. Point zero charge (pH_{ZPC})

One important factor in the adsorption of dyes is the pH of the water. The pH_{ZPC} of ZnO/GO was determined by plotting the initial pH against the variations between the initial and end pH to examine the influence of pH on MG and MO adsorption. The ZPC analysis was conducted using the available criteria in the literature [2,12]. As illustrated in Figure 6(a), the pH_{ZPC} of ZnO/GO was 7.24. The pH_{ZPC} idea states that at low pH, the adsorbent's surface is strongly positively charged, which should cause the cationic dye molecules to repel one another and reduce the efficacy of adsorption for cationic dye. As MO carries a negative charge it can be easily adsorbed at lower pH. On the other hand, the adsorbent surface becomes negatively charged when the solution pH rises over pH_{ZPC} , and electrostatic interactions between the cationic MG and the adsorbent surface should encourage adsorption. The adsorption of MG and MO onto ZnO/GO is possibly controlled by the electrostatic forces of attraction, as shown by Equations (3–6).

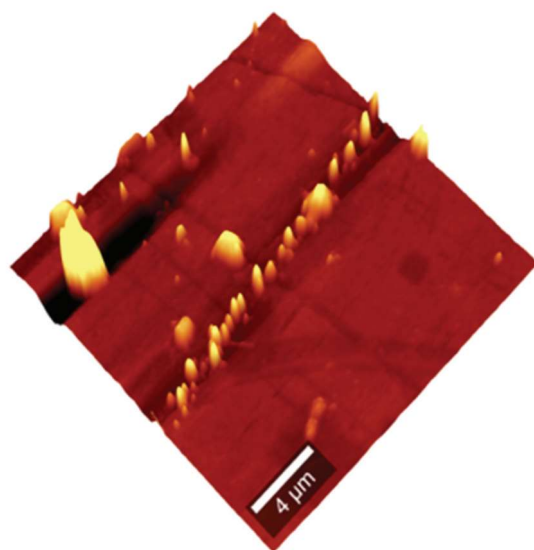


3.1.9. Raman spectroscopy of ZnO/GO

Figure 6(b) depicts Raman spectra of ZnO/GO. Where peaks appearing at ~ 114 and 320 cm^{-1} can be assigned to the E^2 (low) and E^2 (high) phonons of Zn-O bonds respectively. Moreover, the peak at $\sim 560\text{ cm}^{-1}$ corresponds to the A_1 (LO) mode of ZnO particles. Another peak at $\sim 1060\text{ cm}^{-1}$ can be ascribed to the transverse optical and longitudinal optical phonon modes of ZnO. Further, peaks at 1350 and 1595 cm^{-1} correspond respectively to the D and G bands of GO. This suggests the successful formation of the ZnO/GO nanocomposite.



(a)



(b)

Figure 5. (a) 2D AFM image of ZnO/GO. (b) 3D AFM image of ZnO/GO.

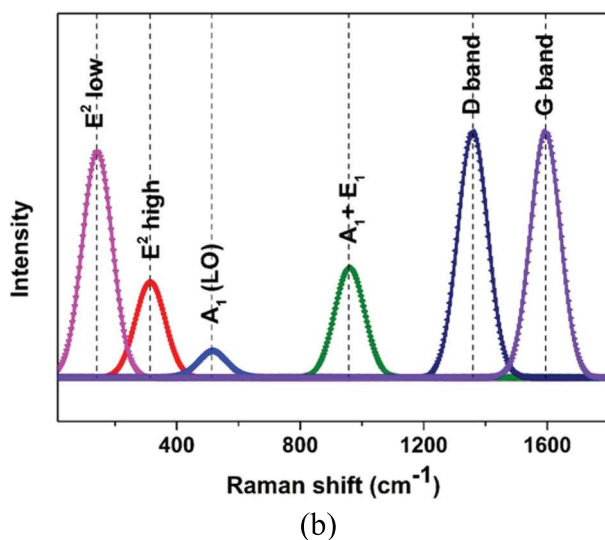
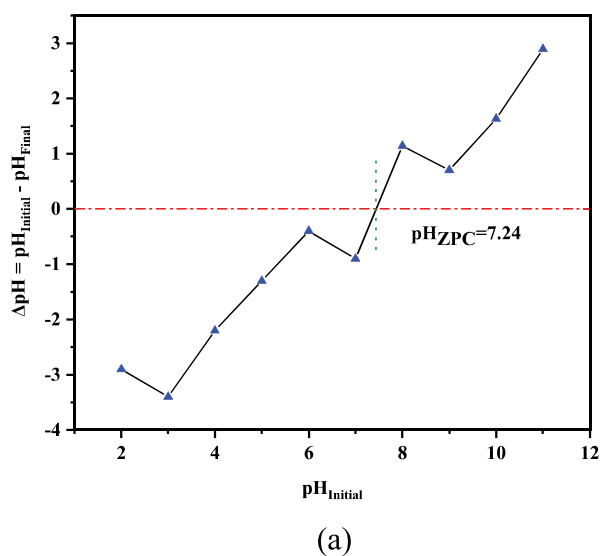


Figure 6. (a) The point zero charge on ZnO/GO. (b) Raman spectra of ZnO/GO.

3.2. Effects of changing operational settings on the capacity of ZnO/GO to adsorb MO and MG

3.2.1. pH

The pH of the solution is a key element that has a substantial impact on adsorption. All the parameters were kept constant while the pH was changed between 2 and 14 to determine its impact on MG and MO adsorption. In the case of MO, adsorption effectiveness improves steadily until pH 4, when pH is increased. At pH 4, the greatest adsorption effectiveness was found. Due to electrostatic attraction, the nano-adsorbent surface becomes protonated at acidic pH, which increases the adsorption of MO's negatively

charged ions. With the increase in pH, the charge present on the surface of the adsorbent is changed to negative from positive (deprotonation), which successively reduces adsorption effectiveness. At higher pH, OH^- ions also have competition with MO's ions having a negative charge, lowering MO's ability to adsorb on the nano-adsorbent surface. Therefore, at pH 4, maximum efficiency was recorded, and as pH rose, efficiency declined. Adsorption efficiency in the MG scenario was found to be fairly low at acidic pH and to be quite high at basic pH. At pH 12, the highest efficiency was seen for the adsorption of MG. Since MG ions have a positive charge, they compete with H^+ ions to be adsorbed at acidic pH. Additionally, the nano-adsorbent's protonated surface prevents adsorption because of repulsive interionic interaction. Deprotonation happens as pH rises, causing higher adsorption of MG on ZnO/GO. Similar outcomes were seen when CV and MO were remedied using *Spirulina platensis* mediated FeO NPs (SP-IONPs) [33]. Figure 7(a) represents the findings.

3.2.2. Nano-adsorbent concentration

One crucial factor that can have an impact on the overall remediation procedure for MO and MG is the concentration of nano adsorbent used. To find the impact of the concentration, the concentration of the nano-adsorbent was adjusted between 0.01 g and 0.05 g during the current study. Up until 0.02 g, the concentration of the nano adsorbent and the adsorption efficiency grew linearly; after that, the adsorption efficiency declined as the concentration increased. Maximum efficiency absorbed at 0.02 g for both dyes. The improved diffusion of ZnO/GO in solution may be the cause of the improvement in remediation at low concentrations of adsorbent. The fact that every active site on the adsorbent surface is fully exposed could speed up dye molecules' approachability to more active sites. Conversely, when the amount of adsorbent increases, a greater proportion of the lower energy active sites occupy and the accessibility of the higher energy adsorbent active sites reduces, which results in a reduction in the adsorption capacity. Furthermore, an overabundance of nanoparticles leads to cluster formation, resulting in larger particles and a reduced surface area, all of which lower ZnO/GO's adsorption efficacy. Comparable findings were seen when palladium and FeO NPs (S-IONPs) derived from *spirulina* were used for the adsorption of lead and CV dye, respectively [3,34]. Figure 7(b) represents the impacts of the concentration of ZnO/GO on the adsorption of MG and MO.

3.2.3. Adsorption isotherm

For both MO and MG, the four other parameters remained unchanged while dye concentrations ranging from 10 to 80 mg/L were used to study the effects. For MG and MO, the highest adsorption effectiveness was attained at 40 mg/L. Thereafter, as the concentration of dye increased, the efficacy of ZnO/GO to adsorb dye gradually declined. The results show that, in both situations, the dye concentration directly influences the efficiency of ZnO/GO. As the concentration of dyes surged, more dyes could be absorbed but active sites on which they have to be adsorbed were limited in number, thus efficiency decreased after all the active sites were occupied [35]. Figure 7(c) represents findings of the impact of the concentration of MO and MG on their adsorption by ZnO/GO.

Adsorption isotherms are mathematical expressions that characterise the dispersal of adsorbate molecules between the adsorbent and liquid based on homogeneity or heterogeneity, the adsorbate molecules' interactions with one another, and the kind of

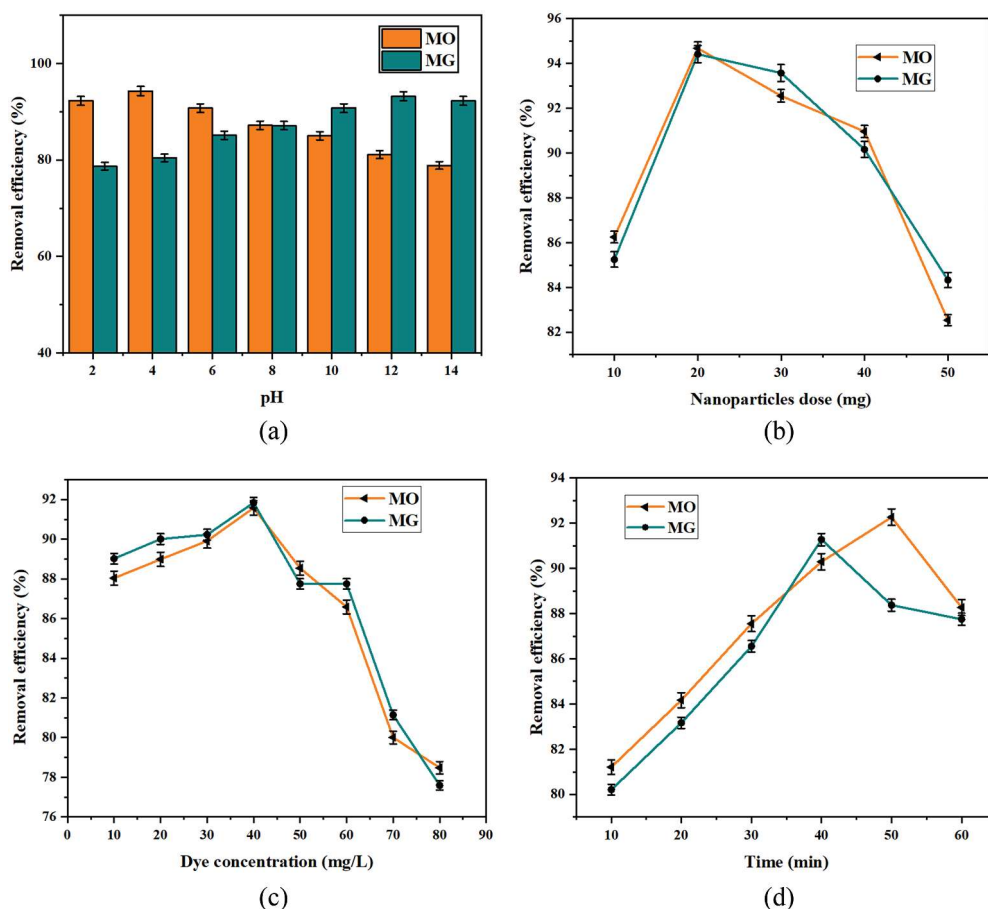


Figure 7. (a) The remediation efficiency of ZnO/GO for MO and MG as a function of pH. (b) The remediation efficiency of ZnO/GO for MO and MG as a function of nanoparticle concentration. (c) The remediation efficiency of ZnO/GO for MO and MG as a function of dye concentration. (d) The remediation efficiency of ZnO/GO for MO and MG as a function of time.

coverage. The Langmuir, Temkin, Redlich Peterson, Dubinin-Radushkevich and Freundlich isotherms were used to investigate the adsorption process further. The Langmuir model postulates that adsorbate molecules will adsorb monolayerly on particular active sites of a homogeneous surface. This model's fundamental presumptions are: (i) Every site has the same energy makeup and can only contain one adsorbed molecule at a time. (ii) The adsorbed molecules on the nearby sites do not combine [36,37]. Equation (S4) represents Langmuir's concept mathematically. Equation (S5) depicts the Langmuir isotherm in its linear form. The dimensionless Langmuir constant, R_L , is used in equation (S6) to show that adsorption can be either linear when R_L equals one, irreversible when R_L is zero or favourable when R_L is greater than 0 but smaller than 1.

The partnership between the concentration of an adsorbate in the solution at equilibrium and the quantities of an adsorbate adsorbed on the surface of the adsorbent is described by the Freundlich isotherm equation. Features of the Freundlich model include

reversible heterogeneous multilayer adsorption [38–40]. The Freundlich model is depicted by equation (S7). Equation (S8) depicts the linear version of the Freundlich isotherm.

Temkin isotherm model primarily assumes that: (i) adsorbate is dispersed over heterogeneous active sites on the adsorbent surface in a monolayer, or (ii) binding energy is distributed uniformly and the heat of adsorption reduces gradually as the result of interaction between adsorbate and adsorbent [41–43]. The mean free energy and adsorbents' characteristics were described using the D-R model for the experimental data. The equation below can represent the Dubinin-Radushkevich isotherms model [42,43]. Because of its versatility, the Redlich-Peterson isotherm model may be applied to both homogeneous and heterogeneous systems, and it is used to depict the equilibrium of adsorption over a wide concentration range [44]. In equations (S9), (S10), and (S11) Temkin, Dubinin-Radushkevich, and Redlich Peterson isotherm's linear form is demonstrated.

The R^2 values for Dubinin-Radushkevich, Temkin, Freundlich, Redlich Peterson, and Langmuir were 0.92492, 0.96009, 0.939, 0.95513, and 0.985621 for MO and 0.92637, 0.94777, 0.9174, 0.95177, and 0.97305 for MG, respectively. The Langmuir isotherm yielded a higher R^2 and lower χ^2 value than other models. It is clear from the research that the Langmuir model matches the current findings. Thus, it indicates that adsorption takes place in a monolayer on consistent surfaces and that there are relatively few interlinkages between the adsorbed species [14,36,37]. Adsorption intensity ($1/n$), which ranges between 0.2 and 0.8, was observed to be 0.6862 and 0.65267 for MO and MG, respectively. The $q_{\max}$ derived using Langmuir was 296.73 mg/g and 259.74 mg/g for MO and MG, respectively. Fig. S3, Fig. S4, and Fig. S5 represent the findings of adsorption isotherms for both MO and MG. Table 3 lists the characteristics of each adsorption isotherm.

3.2.4. Kinetic studies

To examine the effects of time, it was changed from 10 to 60 minutes for both MG and MO. The adsorption efficiency rose from 10 minutes to 50 and 60 minutes, reaching its highest for MO and MG at 50 and 60 minutes, respectively. The accessibility of numerous open sorption sites during the initial stage was the cause of the steady increase in the rate of adsorption. As dye ions accumulated on sorption sites over time, an equilibrium was eventually reached. Due to ZnO/GO reaching a state of equilibrium concerning MO and MG ions after 50 and 60 minutes, adsorption efficiency was raised until that point but did not continue to rise [33,45]. Figure 7(d) illustrates the impacts of time.

The extra dynamics of the process were assessed using adsorption kinetics. The pseudo 1st order, pseudo 2nd order, Elovich, Bangham, and IPDM kinetic models were applied. According to the first kinetic model, which is typically true during the early stages of adsorption, the amount of solid uptake over time and the fluctuation in saturation concentration are closely connected with the rate at which solute uptake changes. The second-order kinetic model states that the rate-limiting phase is chemisorption, and this is true for the entire adsorption extent [23]. Equations (S12) and (S13) display the pseudo-1st linear and non-linear forms respectively and equations (S14) and (S15) display the pseudo-2nd-order linear and non-linear mathematical statements, respectively [2,46].


Table 3. Isotherm parameters for the adsorption of methyl orange and malachite green by ZnO/GO.

Isotherm models	Parameter	MO			MG		
		Value	SE	t-value	Value	SE	t-value
Langmuir	Intercept	0.00337	0.00143	0.95999	0.00385	0.00312	1.537
	Slope	0.04933	0.0038	12.98114	0.04464	0.00311	14.129
	$q_{\max}$ (mg/gm)	296.735905			259.7402597		
	K_L (L mg ⁻¹)	0.068315			0.08624552		
	R_L	0.000677			0.000929		
Freundlich	R^2	0.98562			0.97305		
	χ^2	0.002			0.004		
	Intercept	1.353	0.05539	24.43813	1.49246	0.05962	25.03176
	Slope	0.686	0.07101	9.66313	0.63002	0.07718	8.16307
	$1/n$ (L mg ⁻¹)	0.6862					
Temkin	K_f (mg g ⁻¹)	22.57668					
	R^2	0.939					
	χ^2	0.052			0.9174		
	Intercept	13.33627	5.15195	2.58859	0.045	8.32653	2.59808
	Slope	49.29169	2.86845	17.18406	21.633	4.68097	10.43414
Dubinin-Radushkevich	B (KJmol ⁻¹)	50.26348255			48.84191		
	A (Lg ⁻¹)	1.303858			50.72635366		
	R^2	0.96009			1.531832477		
	χ^2	0.021			0.94777		
	Intercept	4.8679	0.08847	55.02328	0.034	0.08637	56.26559
Redlich Peterson	Slope	-0.000000641678	0.000000074639	-8.59708	4.8598	0.00000005	-8.68865
	R^2	0.92492			-0.0000005	0.00000006	
	χ^2	0.051			0.92637		
	Intercept	-2.79975	0.05383	-52.01313	0.062	0.0491	-63.9656
	Slope	-0.3387	0.02997	-11.30147	-3.14064	0.0276	10.88164
	R^2	0.95513			0.30036		
	χ^2	0.043			0.95177		

Graphic representations of the linear pseudo-first and linear second-order kinetics are depicted in Fig. S6. and Fig. S7. represents non-linear pseudo-first-order and second-order kinetics. It was discovered that the linear pseudo-first-order and linear pseudo 2nd order have R^2 values as 0.95266, 0.99878 for MO and 0.94651, 0.99916 for MG, respectively and non-linear pseudo-first-order and non-linear pseudo-second order have R^2 value as 0.9263, 0.99042 for MO and 0.95401, 0.97736 for MG, respectively. The pseudo-second-order kinetics yielded a higher R^2 and lower χ^2 value than other models. This unquestionably shows that adsorption occurs chemically and follows pseudo-second-order in the cases of MO and MG. K_1 and K_2 , which were calculated to be -0.00004 and 0.008979 for MO and -0.00041 and 0.008509 for MG correspondingly, indicate that the adsorption follows pseudo-2nd order [3,47,48].

Adsorbate is deposited onto the surface of the adsorbent through a multi-step process called (i) adsorbate is transferred from the bulk liquid phase to the surface boundary layer of the adsorbent via bulk diffusion; (ii) film diffusion transfers the adsorbate from the thickness of the boundary layer to the surface of the adsorbent. (iii) adsorbate migration from the surface boundary layer into the interior pore of the adsorbent via intraparticle diffusion, commonly referred to as pore diffusion [33,49]. The intraparticle diffusion model (IPDM) explains the possibilities of intraparticle diffusion. Diffusional mechanisms and surface chemical changes might regulate the general kinetics of sorbate migration onto sorbent surfaces from fluids under investigation. Elovich's theory was extensively explored to investigate the regulating step of sorption. To assess whether pore diffusion predominates in the adsorption process, Bangham's kinetic model is employed [50]. Equations (S16), (S17) and (S18) present the linear form proposed by Elovich, IPDM, and Bangham models respectively.

It is most likely that the outer border coating of the nanoparticles played a major role in the process and that diffusion among the particles was rarely the main regulatory element because it did not pass the origin in the graph (Fig. S8) of the IPDM analysis of MG and MO. The Elovich model's basic postulate is that various active sites of nano-adsorbent fall into a variety of categories and, as a result, have varying levels of adsorption capacity. Using equation (S16), the values for α and β are 0.16951 and 685.501 MO and 0.2681 and 963.732 for MG correspondingly. R^2 values obtained from Elovich, Bangham, and IPDM are 0.97418, 0.98264, and 0.959 for MO and 0.94147, 0.99946, and 0.98675 for MG. Thus, it may be concluded that ZnO/GO has a strong affinity for MO and MG adsorption [33,51,52]. Fig. S8 and Fig. S9 represent the findings of the IPDM, Elovich, and Bangham models. Table 4 indicates the characteristics of adsorption kinetics for both MO and MG.

3.2.5. Thermodynamic studies

To explore the impact of temperature, it was changed between 20°C and 120°C. From 20°C to 60°C, efficiency increased; but, after that point, adsorption efficiency nearly stayed constant. The temperature at which MO and MG were remedied most effectively was 60°C. The obtained results demonstrated that the adsorption is chemical in nature and endothermic. This increase in the percentage of dye removed may be related to the adsorbent surface's activation at higher temperatures and the presence of more active sites. Another possibility is that more MO and MG dye ions are diffusing and travelling faster from the bulk mixture in the direction of the ZnO/GO surface, which increases the quantity of dye molecules that have the energy to react chemically with ZnO/GO at higher temperatures [23]. Fig. S10 represents the impacts of temperature on adsorption of MO and MG on ZnO/GO.

Table 4. Kinetic parameters for adsorption of methyl orange and malachite green by ZnO/GO.

Kinetic models	Parameter	MO			MG		
		Value	SE	t -value	Value	SE	t -value
Pseudo first order	Intercept	2.46864	0.10472	23.57398	2.42815	0.11406	21.2881
	Slope	-0.02413	0.00269	-8.97235	-0.02464	0.00293	-8.4131
	q_e (mg/g)	11.80638			11.33789		
	K_1 (min^{-1})	-0.0004			-0.00041		
	R^2	0.95266			0.94651		
Pseudo second order	χ^2	0.034			0.021		
	Intercept	0.01397	0.00761	1.83439	0.0149	0.00637	2.33973
	Slope	0.0112	0.000195497	57.28391	0.01126	0.00016	68.8581
	q_e (mg/g)	89.28571			88.80995		
	K_2 ($\text{g mg}^{-1} \text{min}^{-1}$)	7971.939			7887.207		
Intraparticle diffusion	R^2	0.008979			0.008509		
	χ^2	0.99878			0.99916		
	Intercept	0.003			0.004		
	K_i ($\text{mg g}^{-1} \text{min}^{-1/2}$)	72.44232	1.39531	51.91855	76.22099	0.489	155.872
	Slope	2.28128	0.23585	9.67257	1.42677	0.08266	17.2616
Elovich	X (mg g^{-1})	2.28128			1.42677		
	R^2	72.44232			76.22099		
	χ^2	0.959			0.98675		
	Intercept	0.043			0.009		
	α ($\text{mg g}^{-1} \text{min}^{-1}$), β (g mg^{-1})	65.41348	1.65794	39.45461	72.24201	1.60542	44.9989
Bangham	R^2	5.89927	0.48021	12.28475	3.72971	0.465	8.02092
	χ^2	0.16951			0.268117		
	Intercept	685.501			963.732		
	Slope	0.97418			0.94147		
	χ^2	0.008			0.042		
Bangham	Intercept	-1.86046	0.01033	-180.06594	-1.8587	0.00174	-1065.62023
	Slope	0.10369	0.00689	15.04816	0.09989	0.00116	85.87249
	R^2	0.98264			0.99946		
	χ^2	0.009			0.008		

The ΔH , ΔS , and ΔG were evaluated to affirm the kind of adsorption of MO and MG on ZnO/GO. Using the Van't Hoff plot, more thermodynamic analysis was conducted. Several thermodynamic research factors are shown in equations (S19), (S20), (S21), and (S22).

The results of the thermodynamic study showed that for MO and MG correspondingly, the values ΔH were 9.35357 KJ/mol and 10.43611 KJ/mol. This positive number denotes an endothermic process. The values of ΔS for MO and MG are 56.1358 J/K/mol and 56.97509 J/K/mol, respectively. The positive value of ΔS represents the fact that randomness increases as adsorption occurs. Also, the positive value of ΔH ΔS both suggests that the reaction is dominated by chemisorption. For both MO and MG, it was found that ΔG was in the negative, which stands for the spontaneous reaction [14,47,53]. Thermodynamic model graphs for MG and MO are depicted in Fig. S11. The thermodynamic model's parameters are shown in Table S1.

3.2.6. Comparison with different adsorbents

To evaluate the performance of ZnO/GO, Table 5 compares ZnO/GO's maximum adsorption capacity with a few other adsorbents that have been discussed in the literature. It is clear from the comparison that ZnO/GO has a good capability for the adsorption of MO and MG [3,14,33,54–56].

Table 5. Comparison of maximum adsorption capacity of ZnO/GO with other adsorbents.

Adsorbent	Dye	Experimental conditions (pH, nanoparticle dose, dye/metal concentration, time, temperature)	$q_{\max}$ (mg/g)	References
S-IONPs	Crystal violet	12, 0.03 g, 20 mg/L, 100 min., 35 °C	55.62	[3]
STONPs	Methyl orange	4, 0.02 g, 30 mg/L, 50 min.	272.47	[14]
STONPs	Malachite green	12, 0.03 g, 30 mg/L, 50 min.	209	[14]
SP-IONPs	Crystal violet	10.4, 0.09 g	256.41	[33]
ZnS:Cu-NP-AC	Malachite green	6, 0.024 g, 10 mg/L, 3 min.	102.60	[54]
ZnAl-LDH	Methyl orange	2, 0.01 g, 30 mg/L, 180 min., 30 °C	86.356	[55]
ZnAl-LDH	Malachite green	10, 0.01 g, 20 mg/L, 180 min., 30 °C	60.606	[55]
ZnO/GO	Methyl orange	4, 0.02 g, 40 mg/L, 50 min., 60 °C	296.73	Present study
ZnO/GO	Malachite green	12, 0.02 g, 40 mg/L, 60 min., 60 °C	259.74	Present study

3.2.7. Mechanism of adsorption

The adsorption of dyes on sorbate is driven by various mechanisms. The process of sorption is greatly influenced by the presence of binding groups on the ZnO/GO surface. The mechanism of adsorption of MO and MG onto ZnO/GO can be easily understood using various forces such as electrostatic interactions, hydrogen bonding and π - π interactions. In the present study, the adsorption of MO and MG perfectly followed the pH_{zpc} theory and thus electrostatic forces are the main forces of attraction between dye ions and the surface of ZnO/GO. The pH_{zpc} of ZnO/GO was 7.24, thus at low pH (<7.24), the ZnO/GO's surface is strongly positively charged, which should encourage the adsorption of anionic MO dye and at higher pH (>7.24), the surface of ZnO/GO's surface negatively charged thus cationic MG dye and easily adsorbed on it. Equations (3-6) described the interactions between the surface of ZnO/GO and both dyes. Characteristic rings of dye interact with the hydroxyl group (OH^-) present on the surface of ZnO/GO through hydrogen bonding. Dye molecules can easily interact with other molecules of dye present in the solution through π - π interactions [57,58]. FTIR of ZnO/GO loaded with MO and MG shows that peaks are shifted and their intensity is changed, again confirming the adsorption of both dyes onto ZnO/GO. Additionally, ZnO/GO's high surface area offers benefits for MO and MG adsorption on its surface. Langmuir adsorption isotherm suited better to adsorption of both dyes thus confirming that adsorption takes place in monolayer on consistent surfaces and that there are relatively few interlinkages between the adsorbed species and according to kinetic studies, chemisorption is the step in the adsorption process that determines rate for both MG and MO, other processes, such as surface adsorption, film diffusion, or external diffusion, may also be at play. The influence of contact time on the adsorption mechanism is characterised by the initial rapid phase of adsorption followed by a slight decrease in the rate of adsorption till equilibrium is attained. Intraparticle and pore diffusion also play an important role during adsorption. Thermodynamic investigations also demonstrated that the adsorption process includes chemisorption. Overall, the adsorption is regulated by electrostatic interactions and chemical reactions that together work for the adsorption of MG and MO on ZnO/GO. Fig. S12 represents the mechanism of adsorption of MG and MO onto ZnO/GO.

3.2.8. Regeneration study of ZnO/GO

Regeneration research was done to determine how long the adsorbent and adsorbate would remain bound to one another. Removable efficiency, stability, and repeatability are important characteristics of an adequate adsorbent from a practical aspect. The present study regenerated dye-loaded ZnO/GO utilising pure ethanol and fine concentration of NaOH in ethanol solution for MO and 0.1 M HCl, 10% acetic acid, and pure acetic acid for MG, since organic solvents may be a promising candidate for dye recovery. The regeneration efficiency increased to over 92% during the first two cycles and remained above 80% during the fourth cycle for MO when the NaOH concentration was increased from 1 to 6% (v/v). In the first cycle, the efficiency of regeneration increased to 78% for MG, and it continued to be over 69% by the fourth cycle. 6% NaOH/ethanol and pure acetic acid were found to be most effectively regenerate ZnO/GO loaded with MO and MG correspondingly. Similar findings were observed during the desorption study of ZnO nanoparticles loaded with methyl orange and amaranth and maghemite nanoparticles loaded with MG [23,59]. These findings show that ZnO/GO are stable and reusable in practical applications, which qualifies them as highly efficient and affordable adsorbent. Figure 8. (a). and Figure 8. (b). represents the regeneration study of ZnO/GO.

4. Cost-effectiveness, challenges, and prospects

Algae are easily available in all types of water bodies and they can be easily cultured, making them a potentially low-cost resource to be utilised for nanomaterials synthesis. Nanoparticles can be easily synthesised using algae. Algal nanoparticles have a high surface area and thus have high adsorption capacity. Algal nanoparticles can be easily regenerated and reused many times, making them cost-effective adsorbents for the removal of dyes from wastewater. Algal nanoparticles can be easily biodegraded, thus reducing disposal costs. Along with these advantages, algal nanoparticles are also highly efficient in the remediation of dyes from wastewater. However, it is important to consider factors such as scalability of production because most of the present studies on algal nanoparticles are conducted under controlled laboratory conditions. It needs to be replicated in the scenarios of the real world to scale up the technology for more promising and significant results. Nanocomposites such as ZnO/GO should be tested for the removal of other potential pollutants that can be present in industrial effluents and real-world wastewater. The long-term stability of nanocomposites such as ZnO/GO which are synthesised using algae needs to be addressed in further research. When algal nanoparticles can remediate a variety of contaminants present in real wastewater then only this technique can be considered as a cost-effective and truly sustainable method for wastewater treatment [1,14,19,60,61].

5. Conclusions

In the contemporary landscape of materials science and environmental sustainability, the synthesis and application of advanced nanocomposites have emerged as a promising avenue for addressing water pollution issues. In the present study, ZnO/GO were successfully applied as adsorbents for the adsorption of MG and MO. They were synthesised using algae and then characterised by XRD, FTIR, UV-VIS, SEM, and BET techniques. At pH 4,

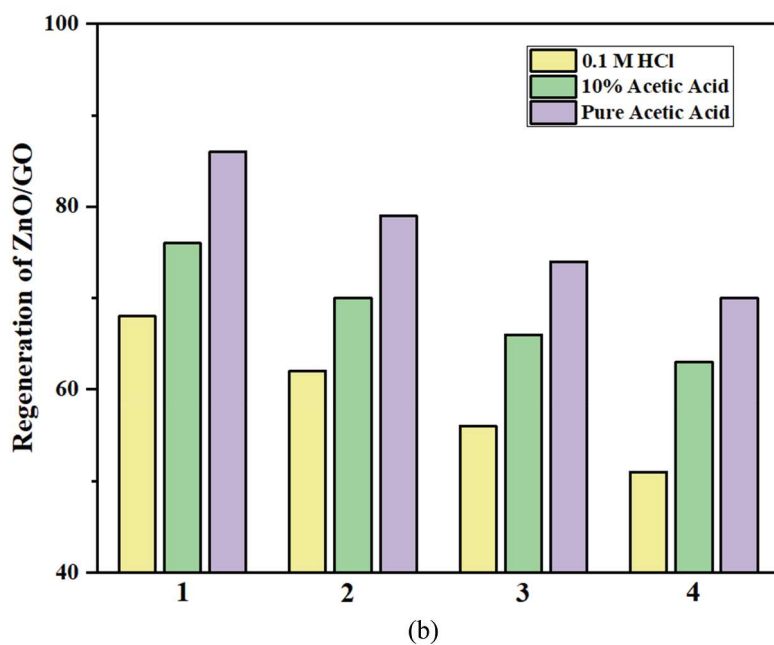
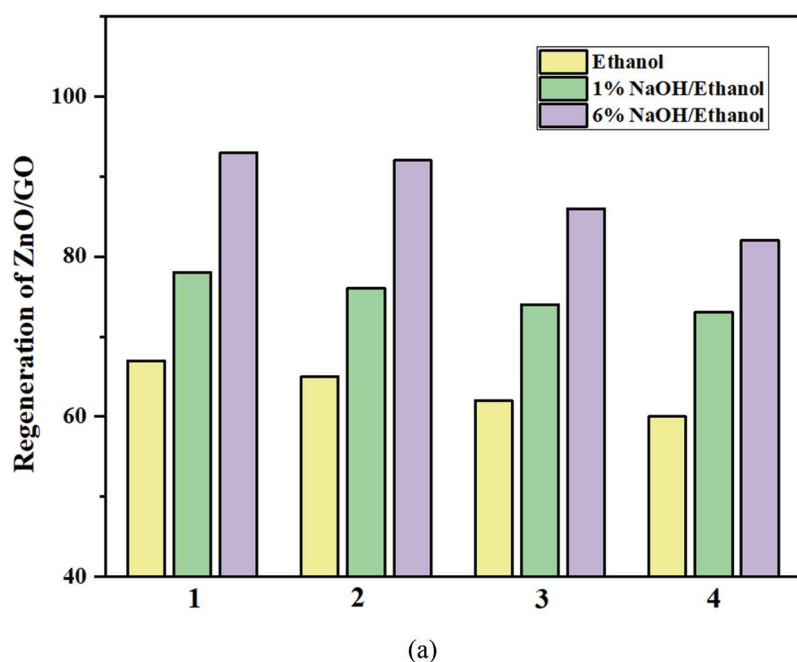


Figure 8. (a) Regeneration of ZnO/GO till 4 cycles for MO. (b) Regeneration of ZnO/GO till 4 cycles for MG.

adsorbent dose of 0.02 g, and temperature 60°C, ZnO/GO removed the most MO, while at pH 12, adsorbent dose of 0.02 g, and temperature 60°C, ZnO/GO removed the most MG. The adsorption investigations verified that the adsorption of MG and MO on ZnO/GO closely adhered to the Langmuir model and the adsorption efficiency was 296.73 mg/g

and 259.74 mg/g for MO and MG. These adsorption processes were found to be pseudo-second-order by means of kinetics studies. The adsorption process's endothermic and spontaneous nature was supported by the thermodynamic studies. The ZnO/GO was found to be reusable till 4 cycles without significant degradation. The scalability of production, long-term stability and its application in real wastewater at a large scale need to be addressed in further research. The synthesis and application of the greenly synthesised ZnO/GO represent a significant step towards sustainable water treatment. The eco-friendly synthesis process, coupled with the exceptional adsorption capacities and reusability of the nanocomposite, positions it as a promising solution for addressing water pollution challenges.

Acknowledgments

One of the authors Anuj Sharma is highly thankful to the All India Council for Technical Education (AICTE) for the AICTE Doctorate Fellowship (ADF).

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

ORCID

Anuj Sharma  <http://orcid.org/0000-0003-4755-4142>
Sharma Mona  <http://orcid.org/0000-0002-9916-1300>
Praveen Sharma  <http://orcid.org/0000-0001-9655-6705>

Data availability statement

All the data generated or analysed during this study are included in this article.

References

- [1] A. Sharma, K. Pal, N. Saini, S. Kumar, D. Bansal and S. Mona, *Nanotechnology* **34**, 352001 (2023). doi:[10.1088/1361-6528/acd45a](https://doi.org/10.1088/1361-6528/acd45a).
- [2] R. Mittal, A. Sharma, A.K. Bhardwaj, R. Bhateria, S. Bansal, R. Kashyap and S. Bhukal, *Inorg. Chem. Commun.* **154**, 110881 (2023). doi:[10.1016/j.inoche.2023.110881](https://doi.org/10.1016/j.inoche.2023.110881).
- [3] S. Bhukal, A. Sharma, D. Rishi, B. Kumar, S. Deepak, S. Pal and K. Mona, *Top. Catal.* **65**, 1675 (2022). doi:[10.1007/s11244-022-01640-3](https://doi.org/10.1007/s11244-022-01640-3).
- [4] M. Lal, P. Sharma and C. Ram, *Results Mater.* **15**, 100308 (2022). doi:[10.1016/j.rinma.2022.100308](https://doi.org/10.1016/j.rinma.2022.100308).
- [5] M. Lal, P. Sharma, L. Singh and C. Ram, *Results Eng.* **17**, 100890 (2023). doi:[10.1016/j.rineng.2023.100890](https://doi.org/10.1016/j.rineng.2023.100890).

- [6] S. Sobhanardakani, R. Zandipak and R. Sahraei, *Toxicol. Environ. Chem.* **95**, 909 (2013). doi:10.1080/02772248.2013.840379.
- [7] S. Sobhanardakani, R. Zandipak, H. Khoshshafar and R. Zandipak, *J. Water Supply Res. Technol. AQUA* **65**, 64 (2016). doi:10.2166/aqua.2016.100.
- [8] S. Sobhanardakani, M. Ghoochian, S. Jameh-Bozorgi and R. Zandipak, *Iran. J. Sci. Technol. Trans. A Sci.* **41**, 1047 (2017). doi:10.1007/s40995-017-0312-z.
- [9] R. Zandipak and S. Sobhanardakani, *Desalin. Water Treat.* **57**, 11348 (2016). doi:10.1080/19443994.2015.1050701.
- [10] B. Saha, S. Shaji and A. Debnath, *J. Dispers. Sci. Technol.* 1–15 (2023).
- [11] S. Patar, S. Konwer, T. Chetia, B.K. Bhuyan and L.J. Borthakur, *Acs Appl. Nano Mater.* **6**, 6567 (2023). doi:10.1021/acsanm.3c00162.
- [12] S. Patar, B. Kumar Bhuyan, M. Konwar, B. Mahanta, P. Saikia, A. Kanti Guha and L. Jyoti Borthakur, *ChemistrySelect* **7** (33) (2022). doi:10.1002/slct.202201936.
- [13] M. Lal, P. Sharma and C. Ram, *Results Opt.* **10**, 100371 (2023). doi:10.1016/j.rio.2023.100371.
- [14] A. Sharma, R. Mittal, P. Sharma, K. Pal and S. Mona, *Nanotechnology* **34**, 485301 (2023). doi:10.1088/1361-6528/acf37e.
- [15] E. Gunasundari, P.S. Kumar, F.C. Christopher, T. Arumugam and A. Saravanan, *IET Nanobiotechnol.* **11**, 754 (2017). doi:10.1049/iet-nbt.2016.0223.
- [16] H. Doshi, A. Ray and I.L. Kothari, *Biotechnol. Bioeng.* **96**, 1051 (2007). doi:10.1002/bit.21190.
- [17] K. Sumit, S. Neha, Sujata, Y. Anjali, S. Anuj, D. Bansal and M. Sharma, in *Integrated Waste Management: The Circular Economy*, edited by H. Diwan and S. Mona (Cambridge Scholars Publishing, UK, 2023), pp. 185–195.
- [18] D.K. Yadav, M. Yadav, R. Mittal, P. Rani, A. Yadav, N.R. Bishnoi and A. Singh, *Sustainable Chem. Environ.* **3**, 100029 (2023). doi:10.1016/j.scenv.2023.100029.
- [19] A. Sharma, P. Sharma and S. Mona, in *Harnessing Microbial Potential for Multifarious Applications*, edited by K. Bala, T. Ghosh, V. Kumar and P. Sangwan (Springer Nature, Singapore, 2024), pp. 359–376. doi:10.1007/978-981-97-1152-9_14.
- [20] M. Kaur, P. Sharma and S. Kumari, *SN Appl. Sci.* **1** (9) (2019). doi:10.1007/s42452-019-1024-0.
- [21] M. Kaur, S. Kumari and P. Sharma, *Biotechnol. Rep.* **25**, e00410 (2020). doi:10.1016/j.btre.2019.e00410.
- [22] S. Patar, R. Mittal, B.K. Bhuyan and L.J. Borthakur, *J. Water Process Eng.* **58**, 104795 (2024). doi:10.1016/j.jwpe.2024.104795.
- [23] M.N. Zafar, Q. Dar, F. Nawaz, M.N. Zafar, M. Iqbal and M.F. Nazar, *J. Mater. Res. Technol.* **8**, 713 (2019). doi:10.1016/j.jmrt.2018.06.002.
- [24] A.A. Hambisa, M.B. Regasa, H.G. Ejigu and C.B. Senbeto, *Appl. Water Sci.* **13** (1) (2023). doi:10.1007/s13201-022-01832-y.
- [25] J. Sharma, S. Sharma and V. Soni, *Reg. Stud. Mar. Sci.* **62**, 102911 (2023). doi:10.1016/j.rsma.2023.102911.
- [26] Y.L. Chen, Z.A. Hu, Y.Q. Chang, H.W. Wang, Z.Y. Zhang, Y.Y. Yang and H.Y. Wu, *J. Phys. Chem. C* **115**, 2563 (2011). doi:10.1021/jp109597n.
- [27] P.K. Narayanam, V.D. Botcha, M. Ghosh and S.S. Major, *Nanotechnology* **30**, 485601 (2019). doi:10.1088/1361-6528/ab3ced.
- [28] V. Yönten, N.K. Sanyürek and M.R. Kivanç, *Surf. Interfaces* **20**, 100529 (2020). doi:10.1016/j.surf.2020.100529.
- [29] S.N. Taqui, C.S. Mohan, B.A. Khatoon, M.E.M. Soudagar, T.M.Y. Khan, M.A. Mujtaba, W. Ahmed, A. Elfakhany, R. Kumar and C.I. Pruncu, *Biomass Convers. Biorefin.* **13**, 9119 (2023). doi:10.1007/s13399-021-01827-w.
- [30] S. Ahmad, A. Ahmad, S. Khan, S. Ahmad, I. Khan, S. Zada and P. Fu, *J. Ind. Eng. Chem.* **72**, 117 (2019). doi:10.1016/j.jiec.2018.12.009.
- [31] Z. Durmus, B.Z. Kurt and A. Durmus, *ChemistrySelect* **4**, 271 (2019). doi:10.1002/slct.201803635.
- [32] S. Víctor-Román, E. García-Bordejé, J. Hernández-Ferrer, J.M. González-Domínguez, A. Ansón-Casaos, A.M.T. Silva, W.K. Maser and A.M. Benito, *Catal. Today* **357**, 350 (2020). doi:10.1016/j.cattod.2019.05.049.

- [33] S.M. Shalaby, F.F. Madkour, H.Y. El-Kassas, A.A. Mohamed and A.M. Elgarahy, *Environ. Sci. Pollut. Res.* **28**, 65549 (2021). doi:10.1007/s11356-021-15544-4.
- [34] M.H. Sayadi, N. Salmani, A. Heidari and M.R. Rezaei, *Surf. Interfaces* **10**, 136 (2018). doi:10.1016/j.surf.2018.01.002.
- [35] J. Hu, G. Chen and I.M.C. Lo, *Water Res.* **39**, 4528 (2005). doi:10.1016/j.watres.2005.05.051.
- [36] P.Z. Ray and H.J. Shipley, *RSC Adv.* **5**, 29885 (2015). doi:10.1039/C5RA02714D.
- [37] P.S. Kumar, R. Gayathri, C. Senthamarai, M. Priyadharshini, P.S.A. Fernando, R. Srinath and V. V. Kumar, *Korean J. Chem. Eng.* **29**, 1752 (2012). doi:10.1007/s11814-012-0077-1.
- [38] P.V. Sierra-Trejo, E. Guibal and J.F. Louvier-Hernández, *J. Polym. Environ.* **28**, 934 (2020). doi:10.1007/s10924-020-01654-6.
- [39] K. Moodley, R. Singh, E. Musapatika, M. Onyango, A. Ochieng, *Warer Sa* **37**, 41–46 (2011). doi:10.4314/wsa.v37i1.64105.
- [40] K.Y. Foo and B.H. Hameed, *Chem. Eng. J.* **156**, 2 (2010). doi:10.1016/j.cej.2009.09.013.
- [41] M. Ngabura, S.A. Hussain, W.A.W.A. Ghani, M.S. Jami and Y.P. Tan, *J. Environ. Chem. Eng.* **6**, 2528 (2018). doi:10.1016/j.jece.2018.03.052.
- [42] A.O. Dada, A.P. Olalekan, A.M. Olatunya and Dada, *IOSR J. Appl. Chem.* **3**, 38–45 (2012). <https://org/www.iosrjournals.org>.
- [43] A.S. Thajeel, *Aquat. Sci. Technol.* **1** (2013). doi:10.5296/ast.v1i2.3890
- [44] J.S. Piccin, G.L. Dotto and L.A.A. Pinto, *Braz. J. Chem. Eng.* **28**, 295 (2011). doi:10.1590/S0104-66322011000200014.
- [45] A. Ayub, A. Irfan, Z.A. Raza, M. Abbas, A. Muhammad, K. Ahmad and A. Munwar, *Polym. Bull.* **79**, 807 (2022). doi:10.1007/s00289-020-03523-7.
- [46] Y.S. Ho and G. McKay, *Process Biochem.* **34**, 451 (1999). doi:10.1016/S0032-9592(98)00112-5.
- [47] S. Mohan, G. Govindankutty, A. Sathish and N. Kamaraj, *Can. J. Chem. Eng.* **99**, 294 (2021). doi:10.1002/cjce.23854.
- [48] A. Ayub, Z.A. Raza, M.I. Majeed, M.R. Tariq and A. Irfan, *Int. J. Biol. Macromol.* **163**, 603 (2020). doi:10.1016/j.ijbiomac.2020.06.287.
- [49] B.C. Melo, F.A.A. Paulino, V.A. Cardoso, A.G.B. Pereira, A.R. Fajardo and F.H.A. Rodrigues, *Carbohydr. Polym.* **181**, 358 (2018). doi:10.1016/j.carbpol.2017.10.079.
- [50] U.A. Edet and A.O. Ifelebuegu, *Processes* **8**, 665 (2020). doi:10.3390/pr8060665.
- [51] R. Foroutan, S.J. Peighambaroust, S.H. Peighambaroust, M. Pateiro and J.M. Lorenzo, *Molecules* **26**, 2241 (2021). doi:10.3390/molecules26082241.
- [52] F. Krika and O.E.F. Benlahbib, *Desalin. Water Treat.* **53**, 3711 (2015). doi:10.1080/19443994.2014.995136.
- [53] A. Saravanan, P.S. Kumar and A.A. Renita, *J. Clean. Prod.* **172**, 92–105 (2018). doi:10.1016/j.jclepro.2017.10.109.
- [54] M. Dastkhoon, M. Ghaedi, A. Asfaram, A. Goudarzi, S.M. Langroodi, I. Tyagi, S. Agarwal and V. K. Gupta, *Sep. Purif. Technol.* **156**, 780 (2015). doi:10.1016/j.seppur.2015.11.001.
- [55] M.A. Nazir, M.A. Bashir, T. Najam, M.S. Javed, S. Suleman, S. Hussain, O.P. Kumar, S.S.A. Shah and A. Ur Rehman, *Microchem. J.* **164**, 105973 (2021). doi:10.1016/j.microc.2021.105973.
- [56] S. Das, S. Rudra Paul and A. Debnath, *J. Mol. Liq.* **387**, 122610 (2023). doi:10.1016/j.molliq.2023.122610.
- [57] P. Das and A. Debnath, *J. Dispers. Sci. Technol.* **44**, 2587 (2023). doi:10.1080/01932691.2022.2110110.
- [58] A. Deb, A. Debnath, K.L. Bhowmik, S. Rudra Paul and B. Saha, *Int. J. Environ. Anal. Chem.* **103**, 5938 (2023). doi:10.1080/03067319.2021.1946683.
- [59] A. Afkhami, R. Moosavi and T. Madrakian, *Talanta* **82**, 785 (2010). doi:10.1016/j.talanta.2010.05.054.
- [60] A. Deb, S. Das and A. Debnath, *Chem. Phys. Lett.* **830**, 140820 (2023). doi:10.1016/j.cplett.2023.140820.
- [61] B. Saha, S. Gayen and A. Debnath, *J. Hazard. Toxic Radioact. Waste* **27** (4) (2023). doi:10.1061/JHTRBP.HZENG-1213.