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Preparation and characterization of an algal-based magnetic biochar nanocomposite for the removal of azocarmine G2 dye from aqueous solutions

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Abstract

Dyes are released into bodies of water as the textile industry expands in response to the growth of the global population. These textile dyes have severe effects on the environment, including wildlife, terrestrial species, and humans. This study explores the synthesis, characterization, and application of an algal-based magnetic biochar nanocomposite for the efficient adsorption of azocarmine G2 (ACG2) dye from aqueous solutions. The magnetic biochar ($Fe_3O_4@BC$) was characterized by Raman spectroscopy, scanning electron microscopy (SEM), X-ray diffraction (XRD), and Fourier transform infrared spectrometry (FTIR). Batch adsorption experiments were performed to assess the impact of the initial dye concentration (25 to 100 mg / L), contact time (up to 300 min), pH (1-3) and temperature (298 to 328 K). The nano-composite achieved a maximum adsorption capacity (q_{max}) of 71.3 mg/g at pH 1, with equilibrium reached within 240 min. Adsorption kinetics followed a pseudo-second-order model (R2 = 0.99), while isotherm analysis fit well with the Langmuir model (R2 = 0.98), indicating monolayer adsorption. However, the Freundlich model provided a better fit, indicating that the multilayer covered a heterogeneous surface with a chemisorption process. The nanocomposite demonstrated as > 90% adsorption efficiency for ACG2 under a variety of conditions, with reusability tests showing retention of over 80% adsorption capacity after five regeneration cycles. This study focusses on the synthesis of an algae-derived biochar with magnetic properties, enhancing its efficiency in post-adsorption separation. The adsorption of Azocarmine G2 (ACG2), a hazardous azo dye, is addressed herein for the first time, establishing the novelty of this research within the domain. Furthermore, this innovative $Fe_3O_4@BC$ adsorbent compound effectively resolves the issue of recyclability. The results highlight that the algal-based magnetic biochar nanocomposite is a viable and sustainable adsorbent, demonstrating exceptional dye adsorption capacity, simplified separation processes, and recyclability. Therefore, it is deemed appropriate for extensive applications in wastewater treatment processes.

Keywords Magnetic biochar, Azocarmine G2, Adsorption, Kinetics, Nanocomposite, Wastewater

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Introduction

The expeditious industrialization and urbanization of recent decades have resulted in the emission of several synthetic organic dyes into natural aquatic systems, thereby presenting a significant environmental challenge [1, 2]. Among these dyes, azo dyes, distinguished by their azo (-N=N-) linkage, display exceptional coloration and are thus widely used [3]. Nevertheless, the structural stability of azo dyes, such as ACG2, impedes their biodegradation and renders them persistent in aquatic ecosystems, eliciting concerns regarding potential environmental contamination and adverse health implications [4]. Azocarmine G dyes have attained prominence because of their widespread application across numerous industries, including textiles, leather, and food processing. These challenges have precipitated substantial research efforts aimed at devising efficient and sustainable methodologies for the adsorption of azo dyes from aqueous milieus [5, 6]. ACG2 is one of the most perilous dyes ubiquitously employed across many essential applications, ranging from critical medical procedures to diverse industrial operations. Azocarmine G dyes are sulfonated anionic dyes with intricate chemical architecture, featuring a structure known formally as 7-Phenyl-5-(4-sulfoanilino) benzo[a]phenazin-7-ium-3-sulfonic acid, is encapsulated within its empirical formula, $C_{28}H_{18}N_3NaO_6S_2$. The chromophoric azo groups undergo reductive cleavage and form highly toxic aromatic amines. Because of the electron-withdrawing groups in azo dyes, biodegradation is difficult. The azo groups undergo reductive cleavage to generate highly hazardous aromatic amines, and biodegradation is difficult because azo dyes contain electron-drawing groups. Furthermore, these reductive cleavage products are highly mutagenic and carcinogenic. Every year 175,000 tons dyes are producing among azo dyes are occupied 60% of dye generation and approximately 15% of azo dyes are released [7] into water bodies. The release of this formidable compound into the environment has the potential to cause severe environmental degradation and pose formidable health risks. Consequently, comprehensive treatment strategies should be rigorously implemented before any discharge to mitigate potentially harmful effects.

Traditional wastewater treatment processes, including membrane filtration, coagulation, chemical precipitation, flocculation, electrochemical removal, solvent extraction, biological treatment, ion exchange, etc.[8–11]. However, these methods have disadvantages i.e., incomplete removal, high energy requirement, production of secondary sludge and are inadequate for effectively addressing the adsorption of ACG2 due to its recalcitrant nature [12–14]. Adsorption is considered an efficient and low-cost technique for adsorption of dyes, organic compounds, and drugs from wastewater effluents. Furthermore, the adsorbents can be regenerated through the desorption process, because the adsorption process is reversible, which means that the adsorbates can be transferred from the adsorbent surface to the solution. However, a few challenges have been discovered, including adsorbent separation after adsorption, desorption, regeneration processes, and adsorbent stability.

Accordingly, an increasing focus has been on investigating novel materials and technologies for the efficient adsorption and elimination of these dyes. Several adsorbents are reported in the literature i.e. bentonite, kaolinite, montmorillonite, zeolites [15, 16], agricultural waste and plant-based adsorbents [17], biopolymers including chitosan, cellulose, lignin, alginate/PVA matrix [18, 19], algae, fungi, and bacterial biomass [20, 21], metal-based and inorganic adsorbents [18, 22, 23], synthetic and polymeric adsorbents [24]and biochar [25]. Linguistic cellulosic materials such as rice husk and rice straw are used as adsorbents, and other applications are organic fertiliser additive, making bricks, and production of bioethanol, biochar, and composites. Among these advanced materials, biochar, a carbonrich porous material derived from biomass pyrolysis, has demonstrated remarkable adsorption capabilities [26]. A substantial array of organic wastes, such as agricultural wastes [17], algae (blue algae, green algae, and Enteromorpha prolifera), biochar-architectured chitosan [24], Poplar wood sawdust [27], oil palm frond [28], acid treated rice straw [29], rice husk [30], cassava peel [31] and Kigelia fibrous [32]can serve as raw materials for the preparation of biochar and magnetic biochar [33]. Biochar is produced via thermochemical processes such as pyrolysis, hydrothermal carbonization, and gasification [34]. The unique structural and surface properties of biochar, such as its high specific surface area, abundant functional groups, and exceptional stability, make it an attractive candidate for adsorption applications. There are several algae that were used as biosorbents, but a limited number of reports can be observed in the literature on algae biochar. The algae have abundant functional groups i.e. -COOH, -NH₂, -OH and -SH groups on algae cell wall provide the binding sites for interaction of dyes. Furthermore, macro algae are available abundantly in the sea and sea-costs. Among Ulva fasciata it is abundantly available in sea and sea cost and it has an ulvan polymer which is responsible for the abundant functional groups. Numerous studies have documented the outstanding adsorption capacity of biochar for various organic contaminants, including dyes. For instance, in their research, Ravindiran et al. [35]

demonstrated the effective adsorption of acid blue 210 &7 dye by biochar derived from sewage sludge, emphasizing the importance of biochar's porous structure and surface functional groups in enhancing adsorption efficiency. Similarly, Zhao et al. [36] investigated the adsorption of Methylene Blue dye using bamboo-derived biochar and highlighted the role of biochar surface chemistry in the adsorption process.

Despite its potential, the practical implementation of pristine biochar in large-scale water treatment projects is significantly hindered by formidable challenges. These challenges manifest as difficulties in separation, low reusability, and limited adsorption capacity, particularly in the case of tenacious dyes like ACG2 [37]. To address these challenges, researchers have diligently developed composite materials by incorporating various functional nanomaterials into biochar matrix [38, 39]. These engineered composites aimed to leverage and integrate the robust characteristics of both biochar and incorporated nanoparticles, significantly enhancing the adsorption capabilities. Among these nanoparticles, iron oxide and zinc oxide are particularly promising candidates [40, 41]. Iron oxide nanoparticles, which are characterized by inherent magnetic properties and an extensive surface area, have consistently demonstrated exceptional potential in adsorption applications [30]. Several studies have highlighted their effectiveness in the adsorption of organic pollutants, illustrating the capability of materials based on iron oxides. However, the application of iron oxide-modified biochar for the adsorption of ACG2 has not yet been explored. This manuscript presents a detailed investigation into the preparation and characterization of an innovative biochar-iron oxide nanocomposite precisely engineered for the effective adsorption of ACG2 dye from aqueous solutions. The detailed synthesis and characterization of the composite are comprehensively discussed, with a focus on its structural and morphological properties. Additionally, this study offers a systematic evaluation of the adsorption performance of the composite, thereby elucidating the factors that influence the adsorption process.

Materials and methods

The Ulva fasciata macroalgae was collected from the Bay of Bengal, Visakhapatnam, India. Sigma-Aldrich, Germany provides analytical–grade ACG2, NaOH, $Fe_2(SO_4)_3$ and $FeSO_4$ ·7 H_2O .

Preparation of biochar from algae

The Ulva fasciata algae were cleansed using distilled water to remove soil and debris, followed by air drying for 48 h. The samples were then ground and subjected to oven drying at 343 K for 24 h. Subsequently, pyrolysis was carried out at 723 K for 2 h under a nitrogen gas flow of 10 mL/min with a heating rate of 283 K/min. The algae biochar was rinsed multiple times with ultrapure water, followed by air drying at 378 K overnight.

Preparation of magnetic biochar (Fe₃O₄@BC) nanocomposites

Magnetic biochar (Fe₃O₄@BC) was prepared as described by Kadimpati et al. [3]. Fe⁺²/Fe⁺³ solution was prepared by mixing a ferric sulfate solution (prepared by dissolving 3.70 g of $Fe_2(SO_4)_3$ in 260 mL of ultrapure water) and a ferrous sulfate solution (obtained by dissolving 4.00 g FeSO₄·7H₂O in 30 mL of ultrapure water) and then heating to 333 K while vigorously stirring. Ten grams of BC powder was gradually added to the prepared Fe⁺²/Fe⁺³ solutions and stirred for one hour. Subsequently, a solution of 5 M sodium hydroxide solution was subsequently added dropwise until reaching pH value between 11 and 12. After stirring the suspension for another hour, it was allowed to settle for a day, followed by filtration and drying at 378 K. The dried matter was then subjected to pyrolysis at 723 K for 2 h under a nitrogen gas flow of 10 mL/min with a heating rate of 283 K/min. The carbonized material was washed thoroughly with ultrapure water and oven-dried overnight at 378 K. The carbonized material was then milled, sieved, and stored for utilization (Fig. S1).

Characterization of the prepared nanocomposites

Different tools were used for the chemical, physical, and morphological characterization of the prepared nanocomposites. X-Ray Diffraction Analysis (XRD) was carried out by (PANanlytical, X'PRT PRO). Using Cu-target with Ni-filtered radiation ($\lambda = 1.542$ A°) and the diffraction angle (2θ) was ranged between 2° and 80° . the morphology of crystals was studied using scanning electron microscopy (SEM), while energy dispersive X-ray spectrometry (EDX) was used for determination of chemical composition. The instrument used was a Jeol JSM 5300 from Japan, in which all samples were placed on stubs and gold-coated to ensure their electrical conductivity and the magnification 2500X was used. Fourier transform infrared (FTIR) spectroscopy measurements were performed using an Agilent Cary 630 instrument in the wavelength range of 400-4000 cm⁻¹. The Raman spectra of the composite samples was conducted using Dispersive Raman microscope (Senterra, Brucker, US) with a laser wavelength of 532 nm at 0.2 mW.

Adsorption of ACG2 dye

A stock solution of ACG2 was prepared by dissolving 1.0 g of ACG2 in 1 L of double-distilled water to achieve a concentration of 1000 mg/L. The stock solution was subsequently diluted to create working solutions with different ACG2 concentrations for adsorption experiments which conducted using the batch equilibrium method. In each trial, 50 mL of the ACG2 solution was mixed with a predetermined dose of the adsorbent material in a sealed container. The mixtures were then agitated in a mechanical shaker to facilitate adsorption. Following agitation, the solutions were centrifuged, and the supernatant was analyzed using a visible-UV spectrophotometer (UV5600, Shanghai Metash Instruments Co., Ltd) to determine the residual ACG concentration. Absorbance measurements were taken at a wavelength of 516 nm, which corresponds to the maximum absorption peak of ACG2. The adsorption capacity (q_t) of the adsorbent at time t was calculated using the following equation:

Adsorption capacity,
$$q_t = \frac{(C_0 - C_t)\nu}{m}$$
 (1)

where C_0 , C_t , are ACG2 concentrations at initial, and at equilibrium time (mg/L); qt is adsorption capacity at time t (mg/g); m is the adsorbent mass (g); v is the solution volume (L).

The adsorption percentage of ACG2 dye from aqueous solution is calculated using the following equation.

$$ACG2 \% \text{ removal} = \frac{C_0 - C_t}{C_0} \times 100$$
 (2)

The pH effect on the adsorption of ACG2 using nanocomposite as an adsorbent was investigated. Specifically, 50 mg of the magnetic biochar was added to 50 mL of a 100 mg/L ACG2 dye solution, at initial pH values of 1, 1.5, 2, and 2.5. pH adjustment was done using 0.1 M HCl and NaOH solutions. The mixture was subsequently stirred at 160 rpm over several time periods (15, 30, 45, 60, 90, 120, 180, and 300 min) at ambient temperature, followed by the collection of samples for analyzing the color of ACG2 dye. An isotherm study was also conducted using different concentrations of ACG2 dye solutions (25, 50, 75, 100, and 150 mg/L). For this purpose, 50 mL of ACG2 solutions with various initial concentrations were agitated with the adsorbent at 160 rpm for 180 min at 298 K. The effects of BC dose and contact time on ACG2 dye adsorption were examined by agitating 50 mL of ACG2 dye solutions with initial concentrations of 75, 100, 150, and 200 mg/L (for both BC and $Fe_3O_4@BC$) with different adsorbent doses (1.5, 2, 2.5, and 3 g/L) for various time intervals (0, 15, 30, 45, 60, 90, 120, 180, and 300 min) at 298 K.

Kinetic modeling, adsorption isotherms, and thermodynamics

The adsorption of ACG2 onto Fe₃O₄@BC was analyzed using pseudo-first-order (PFO), pseudo-second-order (PSO), and intraparticle diffusion (IPD) kinetic models. The linear form of the PFO, PSO and IPD models is as follows (Eq. 3-5):

Pseudo first order : $\ln (q_e - q_t) = \ln q_e - k_{PFO}^t$ (3)

Pseudo Second order
$$\frac{t}{q_t} = \frac{1}{k_{PSO} \times q_e^2} + \frac{1}{q_e}$$
 (4)

Intraparticle diffusion : $q_t = k_{ID} \times t^{\frac{1}{2}} + C$ (5)

In this context, qt and qe represent the quantities of ACG2 adsorbed per unit weight on the Fe₃O₄@BC nanocomposite at a given time 't' and at equilibrium, respectively. The kinetic constants kPFO, kPSO, and kID correspond to the PFO, PSO, and IPD models, respectively, and are determined by evaluating the slopes of the plots of In(qe-qt) versus t, t/qt versus t, and qt versus t^1/2. The adsorption capacities of the nanocomposite for ACG2 were assessed employing the Freundlich and Langmuir isotherm models. The Freundlich model presupposes non-ideal adsorption of ACG2 on heterogeneous surfaces, whereas the Langmuir isotherm presumes monolayer adsorption on the adsorbent surface. The linearized equations of these models are presented as follows (Eq. 6–7):

Freundlich isotherm :
$$\log q_e = \log K_F + \frac{1}{n} \log C_e$$
(6)

where, Ce and qe represent the equilibrium concentration (mg L-1) and equilibrium adsorption capacity (mg g^{-1}) of ACG2, respectively, while $KF[(mgg^{-1})(Lmg^{-1})1/n]$ serves as a constant indicative of the adsorption capacity associated with the adsorption process. The Freundlich model posits that the adsorption of ACG2 occurs on heterogeneous surfaces, reflecting a non-ideal sorption mechanism. The Freundlich constant 'n', indicative of adsorption intensity, is deemed more favorable when values fall within the range of 1 to 10. By examining the slope of the plot of log (qe) versus log (Ce), it is possible to determine the Freundlich parameter values, expressed as 1/n and KF.

Langmuir isotherm :
$$\frac{C_e}{q_e} = \frac{1}{q_m \times K_L} + \frac{C_e}{q_m}$$
 (7)

where, qm represents the maximum adsorption capacity of the Fe₃O₄@BC nanocomposite, while KL denotes the Langmuir equilibrium constant (L mg⁻¹). The Langmuir isotherm presupposes the monolayer adsorption of ACG2 on the adsorbent's surface. The parameters qm and KL can be derived from the slope of the plot of 1/qe against 1/Ce. Furthermore, the thermodynamic parameters of adsorption were evaluated over varying temperatures (298–328 K), encompassing calculations of ΔG (Gibbs free energy, J mol⁻¹), ΔH (enthalpy, kJ mol⁻¹), and ΔS (entropy, J mol⁻¹ K⁻¹) associated with ACG2 adsorption on the adsorbents, as referenced in. [26]:

$$\Delta G^o = -RT \ln K_C \tag{8}$$

$$K_C = \frac{C_S}{C_e} \tag{9}$$

$$\Delta G^o = \Delta H^o - T \Delta S^o \tag{10}$$

$$Log\left(\frac{q_e}{C_e}\right) = \frac{\Delta S^o}{2.303R} - \frac{\Delta H^o}{2.303RT}$$
(11)

where R represents the universal gas constant, T denotes the temperature in Kelvin, CS signifies the concentration of ACG2 on the adsorbent measured in milligrams per gram, and Ce is the equilibrium concentration of ACG2 in milligrams per liter. The values of ΔG° , ΔH° and ΔS° are calculated from slope of Log (ge/Ce) vs. 1/T.

Regeneration and reusability of Fe₃O₄@BC

The regeneration and reusability of $Fe_3O_4@BC$ for the adsorption of ACG2 from water were evaluated. After the adsorption process, the nanocomposites were washed with a desorption medium consisting of 100 mL of 99% ethanol [3] under continuous agitation for a duration of 4 h at a temperature of 298 K. Subsequently, the biochar adsorbents were separated, dried, and reutilized in successive cycles. These adsorption-regeneration cycles were conducted five times utilizing the same regenerated nanocomposite, and the quantity of ACG2 desorbed was determined using Eq. (12).

$$q_{des} = C_{des} \frac{V}{W} \tag{12}$$

where V is the solution volume, q_{des} is the desorption quantity, C_{des} is the desorption concentration, and W is the weight of the nanocomposite.

The desorption ratio was also estimated using Eq. (13).

Desorption ratio

$$= \frac{\text{Amount of ACG2 sorbed}}{\text{Amount of ACG2 desorbed}} \times 100$$
(13)

Results and discussion

Characterization of the synthesized BC and Fe $_3O_4@BC$ XRD

The XRD patterns of BC and Fe₃O₄@BC are presented in Fig. 1a, b. The BC (algal biochar) pattern shows characteristic graphite-like crystalline carbon peaks at $2\theta = 25.9^{\circ}$ and 43.9°. Notably, CaO peaks appear at $2\theta = 31.3^{\circ}$, 36.3°, and 47.3°, while CaCO₃ peaks were evident at $2\theta = 23.0^\circ$, 29.3°, 35.9°, 43.2°, 47.5°, 48.5°, and 59.99°. These results indicate the presence of CaO and CaCO₃ in the BC along with low-intensity graphite-like peaks (Fig. 1a). This proposes BC was carbonated with CO₂ to form CaCO₃. Moisture and CO₂ readily react with CaO to produce $Ca(OH)_2$ and $CaCO_3$, respectively. The CO_2 reacted with CaO and Ca(OH)₂ to form thermodynamically stable $CaCO_3$, as described by Eqs. (4) and (5). CO_2 acts as an activation agent, generating numerous small porous structures in biochar, consistent with the results of Shi et al. [42].

$$CaO(s) + CO_2 \rightarrow CaCO_3(s)$$
 (14)

$$Ca(OH)_2(s) + CO_2 \rightarrow CaCO_3(s) + H_2O$$
 (15)

The Fe₃O₄@BC magnetic nanocomposite XRD pattern shows (Fig. 1b) peaks at 21.1°, 35°, 41.3°, and 50.3°, which are assigned to the crystalline structure of Fe₃O₄@BC (JCPDS 19–0629). The peak at 2θ =35.31° was attributed to the incorporation of Fe₃O₄@BC, while peaks at 2θ =30° and 43° were generated by the interaction of Fe₃O₄@ BC and BC. Comparison with JCPDS card no. 36–1451 suggests a polycrystalline hexagonal phase (wurtzite type), which is consistent with previous research [43, 44].

TEM study of Fe₃O₄@BC nano-composite

TEM images of BC and Fe₃O₄@BC are presented in Fig. 1c, d. The BC surface is rough (Fig. 1c) and the Fe₃O₄@BC nanocomposite, a magnetic component was embedded in the carbon layer (Fig. 1d). The material has an irregular shape (some of the particles appeared as spherical, oval and tetragonal shapes), with the magnetic component distributed unevenly covered by the carbon layer. The Fe₃O₄@BC core had a mean diameter of 8 ± 2.8 nm. The iron oxide particles



Fig. 1 XRD analysis of BC (a), Fe₃O₄@BC (b), structural analysis of the TEM images of BC (c), Fe₃O₄@BC (d) and Fe₃O₄@BC-ACG2 (e)

were embedded in BC matrix was, indicating good mechanical bonding. The dispersion of iron oxide particles was observed in BC was consistent with that of previous studies [45]. Further, after adsorption the TEM image showing that the particles are darkened, and morphology is also changed due to the ACG2 covered on particles (Fig. 1e). Selective area electron diffraction (SAD) analysis identified these two main components, with the iron oxide nanoparticles exhibiting significant crystal lattice planes and the

surrounding biochar being amorphous (Figure S2). The SAD pattern shows clear, sharp diffraction rings, indicating the polycrystalline nature of the iron oxide. The XRD pattern (Fig. 1) confirms that the iron oxide particles were mainly magnetite, with minimal additional phases.

SEM and elemental analysis

SEM micrographs of BC, $Fe_3O_4@BC$ and $Fe_3O_4@BC$ -ACG2 are shown in Fig. 2. EDX and map analyses were used to observe elemental distribution on the surface



Fig. 2 SEM micrographs and EDX-mapping for elements of BC (a), Fe $_3O_4@BC$ (b) and Fe $_3O_4@BC$ -ACG2 (c)

morphology and changes in biochar produced from algae BC and, Fe₃O₄@BC magnetic nanocomposite, as indicated in the EDX analysis in Figure S3. As shown in Fig. 2a, SEM micrographs show that BC produced from marine algae biomass has a rough and porous surface with different pore diameters. It is clearly demonstrated that BC had an uneven shape with a wrinkled surface. EDX-mapping showed that elements C (52 W%), O (23 W%), and Ca (19 W%) were evenly distributed on the surface of BC. It should also be noted that the XRD analysis confirmed the presence of CaO and CaCO₃ particles in the BC composite structure., there are many irregular pores in the biochar, which are facilitated to be incorporated with Fe₃O₄ magnetic nanoparticles, as shown in Fig. 2b. Further, after adsorption of ACG2 the Fe@BC nanocomposite rough and porous surface is covered with a dark matter, probably due to ACG2 adsorption on to the porous-like structure appeared to be filled can be seen in Fig. 2c. EDX analysis showed that carbon and Iron are the most active elements in the Fe@BC nanocomposite and observed the Fe and C are 49 and 29% respectively. The EDX results of BC, Fe₃O₄@BC and Fe₃O₄@BC -ACG2 were depicted in Figure S3. After adsorption of ACG2 the EDX results showed that the increase in weight percent of oxygen and carbon elements whereas Fe is decreased (Figure S3). This may be due to covering of carbon and oxygen species on the iron (Figure S3c). These results are indicated that the adsorption of ACG2 occurred on Fe₃O₄@BC nanocomposite.

Study on functional groups using FTIR

The FTIR spectra of BC and Fe₃O₄@BC are presented in Fig. 3a. The BC spectrum shows peaks at 3421, 1635, 1426, and 1160 cm^{-1} , attributed to the vibration of -OH, C=O, carboxyl O=C-O, and alkoxy C-O groups, respectively. The broad band between 3200 and 3600 cm⁻¹ is likely due to the vibration of the hydroxyl group (⁻OH) of cellulose-derived products. The FTIR results revealed an abundance of oxygen-containing functional groups (C=O, O–H, C-O), which can act as electron donors [46]. The hydroxyl and carboxyl groups on the biochar surface play crucial roles in pollutant adsorption [47]. The Fe₃O₄@BC composite has functional groups (-OH, C=O, Fe-O) that are effective for dye adsorption via chemical bonds [48]. After development of Fe₃O₄@BC composite the hydroxyl functional group at 3421 cm⁻¹ (sharp peak) on biochar is shifted to 3433 cm⁻¹ and formed broad peak (Mosaffa et al. 2024). The carboxylic group peak is also shifted 1635 to 1641 cm⁻¹ (Mosaffa et al. 2025) and C-O groups at 1160 shifted to 1174 cm⁻¹ due to the indicated that the involvement in the Fe₃O₄@BC composite formation



Fig. 3 a FTIR analysis of BC, Fe_3O_4@BC, and Fe_3O_4@BC–ACG2 and (b) Raman analysis of BC and Fe_3O_4@BC

(Fig. 3a). Upon the incorporation of Fe_3O_4 into BC, a new adsorption band appeared at 563 cm⁻¹, attributed to the Fe–O stretching vibration, confirming the successful preparation of the magnetic biochar composite [3].

After adsorption of ACG2 on Fe₃O₄ @BC, the peak of 3433 cm⁻¹ is shifted to 3315 cm⁻¹ and formed broad peak due to the involvement of hydroxyl functional group in the adsorption process. The sharp peak at 1641 cm^{-1} due to the presence of C=C shifted to 1657 cm⁻¹ attributed to the participation in the adsorption process. The bands at 1437 and 864 cm^{-1} correspond to the bending vibration of C-H and the out-of-plane deformation of aromatic CH, respectively after adsorption of ACG2. Similarly, a new peak was observed at around 1117 cm⁻¹ due to C-N on due to azo group of ACG2 on Fe₃O₄ @BC after adsorption process (Fig. 3a). Algal biochar and dye interactions can be explained by the following: (1) cationic dye and negatively charged carboxylate group interaction, (2) coordinate bond between the lone pair electrons of the OH or NH_2 group and the dye, (3) Van der Waals forces between the nonpolar groups of both algal cell and dye, and (4) ion-dipole bond between the dye molecule and the negative dipole end of the carbonyl group. Adsorption can occur via chemical (chemisorption) or physical mechanisms. Chemisorption involves the formation of chemical bonds between the adsorbate

molecule (dye) and the adsorbent surface (biochar). Physical adsorption relies on physical forces like van der Waals forces. A larger surface area can accommodate more adsorbate molecules via physical interactions. The porous structure also enables adsorbate diffusion.

Raman spectra

The Raman spectra of the BC, and $Fe_3O_4@BC$, composites are depicted in Fig. 3b. Prominent peaks appear in the ranges of 1327-1380 cm⁻¹ and 1550-1594 cm⁻¹, assigned to the D and G energy bands, respectively [49]. The D band is associated with sp^3 carbon (C) atom vibrations arising from local disorders and defects, while the G band relates to the tensile vibrations of sp2 atoms in a graphite hexagonal lattice [50]. The ratio of the D and G band intensities (ID/IG) can describe graphite material structural irregularities [51]. The ID/IG ratios for BC and Fe₃O₄@BC were 0.7095 and 1.0372, respectively. A high ID/IG ratio indicates more defects and increased functionalities. The ID/IG ratio for BC/Fe₃O₄ was higher than that for BC, indicating that the BC structure was modified by Fe₃O₄ nanoparticles. This increase in the ID/ IG ratio also suggests a reduction in BC content after the formation of the $Fe_3O_4@BC$ magnetic composite [52].

Determination of texture properties via BET

The textural properties (specific surface area, total pore volume, pore size distribution) were investigated using N₂ adsorption–desorption isotherms. The surface area is a critical indicator of the adsorbent capacity. Results are depicted in Table 1 and supplementary Figure S4(a,b). The BC has a specific surface area of $3.80 \text{ m}^2/\text{g}$ and displays a type-II isotherm, indicating a nonporous structure with abundant micropores and mesopores [53, 54]. This suggests a hierarchical porous structure consisting of micropores and mesopores in the Fe-BCs, as confirmed by the pore size distribution. This structure can minimize the mass transfer resistance and facilitate the transport of reactants to the catalysts [55]. The specific surface areas and total pore volumes of BC and $Fe_3O_4@$ BC were 3.804 m²/g and 0.013 cm³/g and 27.411 m2/g and 0.113 cm³/g, respectively (Table 1). Algal biomassderived biochar is known to have a lower surface area. Surface area is the primary physical factor affecting biochar dye sorption. As shown in Table 1, the BC BET area increased from 3.804 m²/g to 51.92 m²/g after magnetization with iron oxide. The large Fe₃O₄@BC surface area likely provides numerous active adsorption sites. Similar results were observed for the pore volume. Compared with BC (0.015 cm³/g), the Fe₃O₄@BC pore volume (0.153 cm³/g) increased by roughly tenfold, suggesting that Fe introduction potentially enhances Fe₃O₄@BC porosity. The average pore sizes of BC and Fe₃O₄@BC were 8.526 and 5.804 nm, respectively (Table 1). The BC and Fe₃O₄@BC BET surface areas are lower than those of most commercial adsorbents, such as activated carbons (1896 m²/g) [56]. The Fe₃O₄@BC pore diameter range of 3.12–3.6 nm indicates mesoporosity.

Adsorption of ACG2 on BC and Fe₃O₄@BC Effect of contact time on ACG2

The effect of contact time on ACG2 adsorption at concentrations of 100 and 50 mg/L using BC and Fe₃O₄@BC adsorbents with a BC concentration of 2 g/L was examined over a duration of 6 h. Equilibrium was achieved after 220 min, as demonstrated in Supplementary Figure S5a, b. Consequently, an equilibrium time of 4 h was established for all subsequent experiments. Additionally, adsorption studies were conducted over a pH range of 1–10.

The effect of pH on adsorption of ACG2

The initial investigations revealed that ACG2 adsorption was most effective under acidic conditions, with an optimal pH range of 1–2.5, as depicted in Fig. 4. The maximum adsorption capacities (q_e) for various ACG2 concentrations in BC were reported as 5.92, 11.2, 15.584, and 16.28 mg/g for initial ACG2 concentrations of 25, 50, 75, and 100 mg/L, respectively, at pH 1 (Fig. 4b). Figure 4a further indicates that the highest q_e (mg/g) at pH 1 occurred at an initial ACG2 concentration of 100 mg/L.

The point zero charge (pH PZC) is noted at pH 2.1 on Fe₃O₄@BC indicated that the below 2.1 the favorable pH for the adsorption of ACG2 (Supplementary Figure S6). ACG2 is anionic dye and can be exists negative charge and at low pH the positive charges are abundantly available on the Fe₃O₄@BC. The integration of marine algal-based BC with magnetic nano-iron oxide showed an increase in ACG2 adsorption capacity by the double at lower pH, indicating the high activity

Table 1 Textural properties of BC and Fe₃O₄@BC

Samples	Specific surface area (m²/g)	Total pore volume (cm ³ /g)	Micropore volume (cm ³ /g)	Mesopore volume (cm ³ /g)	Pore volume (cm ³ /g)	Pore diameter (nm)
BC	3.804	0.013	0	0.013	0.015	3.618
Fe ₃ O ₄ @BC	51.92	0.113	0	0.113	0.153	3.609



Fig. 4 Influence of initial pH on adsorption efficiency of (a, b) BC and (c, d) Fe₃O₄@BC at different ACG2 concentrations

Fe₃O₄@BC at high acid medium. At elevated pH levels, the neutralization of the positive charges on Fe₃O₄@ BC occurs, accompanied by an increase in surface negative charges. This results in electrostatic repulsion between anionic dye molecules and the negatively charged Fe₃O₄@BC surfaces, leading to a reduction in the adsorption capacity of the Fe₃O₄@BC nanocomposite. Consequently, a favorable pH of 1 is indicated, as similar observations have been reported in the case of Congo red dye adsorption on cationic-amino modified banana leaves [57].

At low pH values, the surface of the magnetic biochar tends to be positively charged due to the protonation of functional groups. This condition enhances the electrostatic attraction between the positively charged surface and the anionic dye ACG2, leading to increased adsorption rates [58]. The maximum adsorption efficiency was observed at pH levels below 2, where nearly 86.8% adsorption was achieved within the first 75 min of contact. Similar findings were reported by Li et al.[59], who noted that acidic conditions significantly improved the adsorption capacity of various adsorbents for anionic dyes due to favorable electrostatic interactions.

Influence of adsorbent dosage

The adsorption of ACG2 onto BC (Fig. 5a, b) and Fe₃O₄@ BC (Fig. 5c, d) was evaluated at various dosages (1, 1.5, 2, 2.5, and 3 g/L) at an equilibrium time of 4 h, temperature of 298 K, and pH of 1. The results presented in Fig. 5 indicate a clear trend in the relationship between the adsorbent dosage and the adsorption efficiency of the dye from aqueous solutions. In the case of modified BC with magnetic iron oxide (Fe₃O₄@BC), the adsorption capacities increased to 8.97, 19.94, 29.025, and 39.35 mg/g at 25, 50, 75, and 100 mg/L of ACG2 (Fig. 5c, d). The results revealed that at low dosages (1 and 1.5 g/L), the adsorption efficiency of different was in the range of only 35–55%, for BC and 70–78% for Fe₃O₄@BC, indicating that there were insufficient active sites on the magnetic biochar to effectively adsorb ACG2. However,



Fig. 5 Influence of initial ACG2 concentrations on adsorption efficiency of (**a**, **b**) BC and (**c**, **d**) $\text{Fe}_3\text{O}_4@BC$ at 2 g/L dosage

these small doses showed high q_e values, which reached 23.8 mg/g for BC and 71.3 mg/g for Fe₃O₄@BCat at an initial dye concentration of 100 mg/L. The maximum adsorption efficiency was achieved when 2.5 and 3 g/L of Fe₃O₄@BC were used, with adsorption efficiencies ranging from 91 to 95% at different ACG2 concentrations (Fig. 5d). However, the absorption capacities at these high doses showed lower values (27.1-36.9 mg/g)at different dye concentrations (Fig. 5c). When evaluating the adsorption of ACG2 onto Fe₃O₄@BC, it is essential to understand the dynamics at play when high concentrations of the adsorbent are used. The observed low adsorption capacity coupled with high adsorption efficiency at elevated Fe₃O₄@BC dosages can be attributed to several interrelated factors. At higher doses of magnetic Fe₃O₄@BC, the total number of available adsorption sites increased, which initially enhanced the adsorption efficiency. However, as the concentration of biochar increases beyond an optimal point, many of these sites become saturated with dye molecules [60]. This saturation leads to a plateau in the adsorption capacity, where the additional biochar does not correspondingly increase the amount of dye adsorbed. For instance, studies have shown that once a certain adsorbent threshold is reached, the efficiency may stabilize or even decline due to saturation effects [28]. The diffusion rate of dye molecules toward the adsorbent surface may slow down as more particles are present in solution, creating a barrier for effective adsorption. This phenomenon can lead to a situation in which despite high adsorption efficiencies being recorded (indicating many molecules have been removed from solution), the actual amount adsorbed per gram of biochar is lower than expected due to these kinetic limitations [61].

Impact of temperature on ACG2 adsorption

The impact of temperature on the adsorption of ACG2 using BC and $Fe_3O_4@BCat$ a concentration of 2.5 g/L and pH 1 was assessed at various initial dye concentrations, as shown in Supplementary Figure S7. The results

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indicated that increasing the temperature from 298 to 328 K had a negligible effect on the adsorption capacity of BC and Fe₂O₄@BC. At temperatures of 298, 308, 318, and 328 K and ACG2 concentrations of 25, 50, 75, and 100 mg/L, qe values ranges were 8.9-9.2 mg/g, 17.3-18.2 mg/g, 25.5-27.3 mg/g and 33.8-36.8 mg/g, respectively. The corresponding adsorption efficiencies were 91-95%, 90-95%, 88-94.5% and 86-93%. This low effect of temperature on the adsorption capacity can be attributed to several interrelated factors, including the nature of adsorption interactions (physisorption vs. chemisorption), compensatory mechanisms at play during kinetic energy increases, limited influence on microporous structures, and specific experimental conditions or models used for analysis. Physisorption, which is characterized by weak van der Waals forces, may show less sensitivity to temperature changes compared to chemisorption, which involves `stronger chemical bonds [62]. In cases in which physisorption dominates, the increase in kinetic energy at higher temperatures does not significantly enhance desorption rates, leading to a stable adsorption capacity across a range of temperatures [15]. In the adsorption process using porous materials like magnetic biochar, micropore filling is essential. Temperature minimally affects the microporous volume and structure, keeping the adsorption capacity stable because of the unchanged pore structures [63]. Functional groups on magnetic biochar, such as -OH and -COOH, help offset temperature effects by sustaining dye molecule affinity, stabilizing adsorption performance [64].

Adsorption kinetics, isotherms and thermodynamics

The adsorption kinetics were modeled using pseudo-first-order (PFO) and pseudo-second-order (PSO), as shown in Fig. 6. The kinetic parameters of PFO for various amounts of Fe₃O₄@BC were assessed based on the respective graphs (Fig. 6a). The calculated q_e values were lower than the experimental qe values, indicating that the PFO model does not accurately predict the adsorption



Fig. 6 Results of kinetic study for adsorption of ACG2 on Fe₃O₄@BC: pseudo first-order kinetics for (**a**); pseudo second-order kinetics for (**b**); experimental q_e (**c**) and intra particle diffusion kinetics (**d**)

kinetics of ACG2 on Fe₃O₄@BC. The PSO model provided the best fit for AGC2 adsorption, with an R² value of 0.999 (Fig. 6b). The calculated qe values were very close to the experimental values, and the kinetic experimental data were effectively interpreted by the PSO (Table 2); and experimental q_e (Fig. 6c) are depicted and this conclusion is consistent with previous studies [65, 66] on modified biochar and seaweed biochar for dye sequestration. The plots in Fig. 6d show that the adsorption of ACG2 on Fe₃O₄@BC versus t^{1/2} did not form linear plots and did not pass through the origin. This conclusion confirmed that the rate-controlling step did not involve intra-particle diffusion; this observation aligns with those reported in the literature [65, 67].

The adsorption data of ACG2 by BC and Fe₃O₄@BC were simulated and fitted to both Langmuir and Freundlich isotherm models (Fig. 7). The corresponding isotherm parameters and their correlation coefficients (\mathbb{R}^2) are presented in Table 3. The Freundlich constant K_f decreased with increasing temperature for both BC (Fig. 7a) and Fe₃O₄@BC (Fig. 7b), suggesting enhanced adsorption capacity at room temperature. Furthermore, the Freundlich exponent n exceeds 1, signifying a favorable adsorption process. The superior fit of the Langmuir isotherm suggests that the adsorption behavior is predominantly monolayer, involving both chemical and physical adsorption mechanisms. These findings are consistent with previous reports on azo-dye adsorption by modified biochar [65].

The data exhibit excellent agreement with the Langmuir isotherm, with R^2 values ranging from 0.954 to 0.979 for BC (Fig. 7c) and 0.987 to 0.996 for Fe₃O₄@BC (Fig. 7d). The calculated RL values fell within the range of 0.109–0.175 across the entire temperature study (298– 328 K), indicating favorable adsorption conditions for both BC and Fe₃O₄@BC. The Freundlich isotherm also provides a reasonable fit, although the R^2 values are lower than those obtained with the Langmuir model, as shown in Figs. 7a and b. the R^2 values for BC ranges were 0.907–0.95 while it was 0.977-0.998 for Fe₃O₄@BC.

Gibbs free energy ΔG serves as the thermodynamic criterion at constant pressure (P) and temperature (T) for determining whether a chemical process can occur or proceed. The enthalpy changes (ΔH) and entropy changes (Δ S) for the adsorption process of ACG2 on BC and $Fe_3O_4@BC$ were derived from the plot of ln b versus $1/T^{-1}$, compiled in Table 4, and illustrated Supplementary Figure S 8. The spontaneity of in the reaction can be evaluated by examining the sign and magnitude of ΔG° . A negative ΔG° signifies the spontaneity of a chemical process. In designing any chemical process system, it is crucial to comprehend the changes expected during a chemical reaction. The rate and magnitude of these changes provide valuable insights for designing process equipment. An extensively negative value for ΔG° suggests the spontaneity of the adsorption process at a specific temperature (Figure S8b). With increasing temperature, the free energy values exhibit a positive trend, indicating a reduction in the spontaneity of the adsorption process at elevated temperatures. Positive ΔH° values denote the endothermic nature of the adsorption. The positive values of ΔS° indicate an increase in randomness at the solid/solution interface during the adsorption of ACG2 onto BC Fe₃O₄@BC.

Adsorption mechanism and comparison with other studies

The Fe₃O₄@BC composite exhibits a high surface-tovolume ratio characterized by the presence of pores, pits, holes, and surface defects, as indicated in Table 1. These structural features enabled the composite to adsorb the investigated ACG2 dye via physical and/or chemical mechanisms. The adsorption of dyes can occur within the interstitial spaces or defects among the carbon nanoclusters. Furthermore, additional physical adsorption mechanisms, including pore diffusion, van der Waals forces, π - π interactions, steric effects, and hydrophobichydrophobic interactions, may also facilitate dye uptake

Table 2 Kinetic parameters of the pseudo-first, pseudo-second order kinetic models and intra-particle diffusion at pH = 1, T = 298 K, and an initial ACG2 concentration of 100 mg/L for BC and Fe₃O₄@BC, respectively

Kinetic model	Biochar	$q_e (mg g^{-1})$	k ₁ (min ⁻¹)	Experimental q _e (mg g [−]	⁻¹) r ²
Pseudo-first-order	BC	13.4	0.018	12.4	0.988
	Fe ₃ O ₄ @BC	45.4	0.004	36.5	0.979
Pseudo-second-order	BC	24.5	0.023	18.5	0.988
	Fe ₃ O ₄ @BC	58.8	0.0026	51.2	0.999
Intra-particle diffusion		k_P (mg h ^{0.5} g ⁻¹)	С		r ²
	BC	0.7	27.26		0.856
	Fe ₃ O ₄ @BC	1.516	36.24		0.885

 K_p intraparticle diffusion rate constant, C constant gained from the intercept, r^2 correlation coefficient



Fig. 7 Freundlich isotherm for BC (a) and Fe₃O₄@BC (b), Langmuir isotherms for BC (c) and Fe₃O₄@BC (d) at different temperatures

Table 3 Langmuir and Freundlich models of ACG2 adsorption isotherms on BC and Fe₃O₄@BC at different temperatures

Adsorbent		Langmuir model				Freundlich model		
	Т (К)	<i>q_m</i> (mg/g)	<i>b</i> (L/mg)	RL	R ²	K _f [(mgg ⁻¹) (Lmg ⁻¹) ^{1/n}]	n _f	R ²
BC	298	28.57	0.0483	0.175	0.979	0.4074	1.7921	0.95
	308	30.30	0.0498	0.170	0.954	0.4009	1.7301	0.919
	318	30.30	0.0678	0.131	0.973	0.2917	1.9194	0.946
	328	29.41	0.0840	0.109	0.973	0.2624	1.9920	0.907
Fe ₃ O ₄ @BC	298	71.4	0.0700	0.1275	0.986	0.1786	1.425	0.998
	308	66.7	0.1049	0.0888	0.996	0.1419	1.435	0.988
	318	76.9	0.1048	0.0889	0.981	0.1291	1.348	0.984
	328	83.3	0.1188	0.0792	0.978	0.1205	1.233	0.974

[68]. Furthermore, chemical adsorption pathways involving electron sharing between the dye molecules and the composite surface may occur. In this instance, an elevation in the solution's pH led to the neutralization of

		5 1		
Temp. (K)	$C_0^{}$ (mg L ⁻¹)	ΔH° (kJ mol ⁻¹)	ΔS° (J mol ⁻¹ K)	ΔG° (kJ mol ⁻¹)
298	24.3	27.208	118.635	- 35.326
308	48.1	11.773	62.745	- 19.313
318	72.4	11.248	59.490	- 18.906
328	97.8	4.832	35.288	- 11.569
298	24.3	594.576	2073.635	- 617.349
308	48.1	118.942	444.788	- 136.876
318	72.4	72.433	279.165	- 88.702
328	97.8	38.332	161.142	- 52.816
	Temp. (K) 298 308 318 328 298 308 318 328 318 328	Temp. (K) C ₀ (mg L ⁻¹) 298 24.3 308 48.1 318 72.4 328 97.8 298 24.3 308 48.1 318 72.4 328 97.8 308 48.1 318 72.4 328 97.8	Temp. (K)C₀ (mg L ⁻¹)ΔH° (kJ mol ⁻¹)29824.327.20830848.111.77331872.411.24832897.84.83229824.3594.57630848.1118.94231872.472.43332897.838.332	Temp. (k)C₀ (mg L⁻¹)ΔH° (kJ mol⁻¹)ΔS° (J mol⁻¹ K)29824.327.208118.63530848.111.77362.74531872.411.24859.49032897.84.83235.28829824.3594.5762073.63530848.1118.942444.78831872.472.433279.16532897.838.332161.142

Table 4 Thermodynamic parameters for ACG2 biosorption onto BC and Fe₃O₄@BC

positive charges on Fe₃O₄@BC, along with an augmentation in surface negative charges which induced electrostatic repulsion between anionic dye molecules and the negatively charged Fe₃O₄@BC surfaces. This phenomenon resulted in a reduced adsorption potential of the Fe₃O₄@BC nanocomposite. Consequently, the optimal pH is identified as 1, a conclusion that is also supported by findings concerning the adsorption of Congo Red on cationic-amino modified banana leaves [57].

In recent years, magnetic biochar nanocomposites have been widely used in the field of dye removal. Algal-based magnetic biochar proved to be an efficient adsorbent for ACG2 adsorption. Compared with other studies, the maximum adsorption of similar dyes on other magnetic biochar at the same temperature was predicted through extrapolation of the curves. Biochar prepared from bamboo sawdust through a hydrothermal carbonization method shows a maximum ACG2 adsorption of 49.93 mg/g at 298 K [69], leading to a similar adsorption capacity in this research, which is 54.8 mg/g at 298 K. Therefore, algal-based magnetic biochar can be efficiently and eco-friendly used in dye wastewater treatment.

Reusability of Fe₃O₄@BC composite

The ACG2-loaded $Fe_3O_4@BC$ composites were regenerated via treatment with anhydrous ethanol, followed by filtration and drying at 373 K for 12 h. The recovered $Fe_3O_4@BC$ composite was reused for five consecutive cycles for ACG2 adsorption. Results indicate that the percentage of ACG2 adsorption decreased from 96 to 83% over the course of five cycles (Supplementary Figure S9). The observed reduction in adsorption efficiency after regeneration can be attributed to weight loss during the adsorbent recovery process. Nevertheless, the observed loss is acceptable for the adsorption and recovery of ACG2.

Conclusions

This study conducted a comprehensive investigation into the adsorption of Azocarmine G2 onto marine algal-based magnetic biochar (Fe₃O₄@BC), with a focus on various factors influencing the adsorption process. These factors include pH, adsorbent dosage, initial dye concentration, temperature effects, kinetics, isotherm modeling, adsorption mechanisms, and regeneration potential. The results indicate that the adsorption capacity (qe) of Fe₃O₄@BC for ACG2 is markedly influenced by the pH level. A maximum adsorption efficiency of 96% was observed at a low pH (approximately 1 and qe around 46 mg/g), which was attributed to enhanced electrostatic interactions between the positively charged surface of the biochar and the negatively charged dye molecules. An increase in the dosage of Fe3O4@BC improved the adsorption efficiency of ACG2 to 95.8%, peaking at an optimal point (2.5 g/L), beyond which only a slight increase in efficiency was noted, with the maximum qe reaching 71.3 mg/g. Kinetic analysis revealed that adsorption followed pseudo-second-order kinetics, implying a significant role of chemisorption in dye uptake. Isotherm studies demonstrated that both the Langmuir and Freundlich models could describe the adsorption behavior; however, the Langmuir model provided a superior fit, indicating monolayer coverage on a homogeneous surface. This finding suggests that magnetic biochar possesses a finite number of uniform sites for dye adsorption. Regeneration studies confirmed that magnetic biochar Fe3O4@BC could be effectively reused post-saturation, maintaining a significant portion of its initial adsorption capacity across multiple cycles. The algae biomass is sustainable and abundantly available in nature, and its utilization in biochar production, along with recyclability and regeneration, aligns with the SDG developmental goals, rendering this adsorbent highly suitable for large-scale wastewater treatment applications.

Supplementary Information

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Supplementary material 1.

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Author contributions

Mohamed Hellal: Conceptualization, Visualization, investigation Methodology, Writing- Original draft preparation. Sayed K. Attia& Anna Genida: Methodology, Visualization, and Supervision. Kishore K. Kadimpati: Conceptualization, Visualization Investigation, Methodology, Writing- Reviewing and Editing. Ahmed M. Rashad: Conceptualization, Visualization, Methodology Writing, reviewing, and editing.

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Availability of data and materials

All authors ensure that all data, materials, software applications, and custom code support their published claims and comply with domain standards. The datasets used or analyzed during the current study are available from the corresponding author upon reasonable request.

Declarations

Ethics approval and consent to participate

No specific permissions were required for the collection of macroalgae in this study, as the species sampled are neither endangered nor protected, and the collection was conducted in unrestricted coastal areas in compliance with local regulations.

Consent for publication

Not applicable.

Competing interests

The authors declare no competing interests.

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References

- Hellal MS, Doma HS, Abou-Taleb EM. Techno-economic evaluation of electrocoagulation for cattle slaughterhouse wastewater treatment using aluminum electrodes in batch and continuous experiment. Sustain Environ Res. 2023;33:2.
- Abou-Taleb EM, Ali ME, Hellal MS, Kamal KH, Abdel Moniem SM, Ammar NS, et al. Sustainable solutions for urban wastewater management and remediation. Egypt J Chem. 2020;63:405.
- Kadimpati KK, Gnida A, Turek-Szytow J, Hellal MS, Gregor M, Matula G, et al. Design of innovative hybrid biochar prepared from marine algae and magnetite: insights into adsorption performance and mechanism. Chem Eng Res Des. 2024;201:218–27.

- Kamal KH, Azab El-Liethy MA, Hemdan BA, Hellal MS, Abou-Taleb EM, El-Taweel GE. Impact of discharged textile dyes on environmental water bodies: a physicochemical, eco-toxicological and microbiological assessment. Egypt J Chem. 2024;67:601.
- Prinith NS, Manjunatha JG. Voltammetric analysis of hazardous azo-dye indigo carmine at simple and cost-effective carbon paste sensor modified with L-phenylalanine. Sens Technol. 2023. https://doi.org/10.1080/ 28361466.2023.2218877.
- Senanayake G, Perera P, Attanayake RN. Textile dye decolorization ability of decaying hardwood associated fungi. Proc Int For Environ Symp. 2022. https://doi.org/10.31357/fesympo.v26.5734.
- Thangaraj S, Bankole PO, Sadasivam SK. Microbial degradation of azo dyes by textile effluent adapted, Enterobacter hormaechei under microaerophilic condition. Microbiol Res. 2021;250:126805.
- Hellal MS, Gamon F, Cema G, Hassan GK, Mohamed GG, Ziembińska-Buczyńska A. Nanoparticle-assisted biohydrogen production from pretreated food industry wastewater sludge: microbial community shifts in batch and continuous processes. Energy Convers Manag. 2023;2024(299):117824.
- Al-Sayed A, Hellal MS, Al-Shemy MT, Hassan GK. Performance evaluation of submerged membrane bioreactor for municipal wastewater treatment: experimental study and model validation with GPS-X software simulator. Water Environ J. 2023. https://doi.org/10.1111/wej.12852.
- Hellal MS, El-Kamah HM, Doma HS. High-performance internal circulation anaerobic granular sludge reactor for cattle slaughterhouse wastewater treatment and simultaneous biogas production. BMC Biotechnol. 2024. https://doi.org/10.1186/s12896-024-00849-2.
- 11. Hellal MS, Hemdan BA, Youssef M, El-Taweel GE, Abou Taleb EM. Novel electro-oxidation unit for electro-disinfection of *E. coli* and some waterborne pathogens during wastewater treatment: batch and continuous experiments. Sci Rep. 2022;12:16417.
- Correa-Mahecha F, Cuesta-Parra DM, Perilla AXM, Barriga LAM, Ortiz AMG. Removal of azo dyes through a natural coagulant obtained from coffee waste (Coffea Arabica). Chem Eng Trans. 2023;99:103–8.
- Bourechech Z, Seghier A, Mokhtar A. Assessment of physically treated banana leaves as a low-cost and eco-friendly adsorbent for removal of a textile azo dye. Biomass Convers Biorefinery. 2023;13:14241–52.
- Chafi M, Gourich B, Essadki AH, Vial C, Fabregat A. Comparison of electrocoagulation using iron and aluminium electrodes with chemical coagulation for the removal of a highly soluble acid dye. Desalination. 2011;281:285–92.
- Hellal MS, Rashad AM, Kadimpati KK, Attia SK, Fawzy ME. Adsorption characteristics of nickel (II) from aqueous solutions by Zeolite Scony Mobile-5 (ZSM-5) incorporated in sodium alginate beads. Sci Rep. 2023;13:19601.
- Ranjbari S, Ayati A, Niknam Shahrak M, Tanhaei B, Hamidi TS. Design of [Bmlm]3PW12O40lonic liquid encapsulated-ZIF-8 nanocomposite for cationic dye adsorptive removal: modeling by response surface methodology. Ind Eng Chem Res. 2023;62:4636–45.
- 17. Zahir A, Aslam Z, Aslam U, Abdullah A, Ali R, Bello MM. Paspalum notatum grass-waste-based adsorbent for rhodamine B removal from polluted water. Chem Biochem Eng Q. 2020;34:93–104.
- Kumar KK, Sujatha S, Skarka W, Monfort O. An innovative hybrid biosorbent composed of nano ZnO and marine macro algae Jania rubens embedded in an alginate/PVA matrix: insights into Pb2+ removal in water. New J Chem. 2022;47:373–83.
- Khawar A, Aslam Z, Zahir A, Akbar I, Abbas A. Synthesis of Femur extracted hydroxyapatite reinforced nanocomposite and its application for Pb(II) ions abatement from aqueous phase. Int J Biol Macromol. 2019;122:667–76.
- Sarada B, Prasad MK, Kumar KK, Ramachandra Murthy CV. Cadmium removal by macro algae Caulerpa fastigiata: characterization, kinetic, isotherm and thermodynamic studies. J Environ Chem Eng. 2014;2:1533–42.
- Kadimpati KK, Mondithoka KP, Bheemaraju S, Challa VRM. Entrapment of marine microalga, Isochrysis galbana, for biosorption of Cr(III) from aqueous solution: isotherms and spectroscopic characterization. Appl Water Sci. 2013;3:85–92.
- 22. Shahinpour A, Tanhaei B, Ayati A, Beiki H, Sillanpää M. Binary dyes adsorption onto novel designed magnetic clay-biopolymer hydrogel involves characterization and adsorption performance: kinetic, equilibrium, thermodynamic, and adsorption mechanism. J Mol Liq. 2022;366:120303.

- 23. Pang S, Zhou C, Sun Y, Zhang K, Ye W, Zhao X, et al. Natural wood-derived charcoal embedded with bimetallic iron/cobalt sites to promote cipro-floxacin degradation. J Clean Prod. 2023;414:137569.
- Mosaffa E, Ramsheh NA, Banerjee A, Ghafuri H. Bacterial cellulose microfilament biochar-architectured chitosan/polyethyleneimine beads for enhanced tetracycline and metronidazole adsorption. Int J Biol Macromol. 2024;273:132953.
- Mosaffa E, Banerjee A, Ghafuri H. Sustainable high-efficiency removal of cationic and anionic dyes using new super adsorbent biochar: performance, isotherm, kinetic and thermodynamic evaluation. Environ Sci Water Res Technol. 2023;9:2643–63.
- Praveen S, Jegan J, Bhagavathi Pushpa T, Gokulan R, Bulgariu L. Biochar for removal of dyes in contaminated water: an overview. Biochar. 2022. https://doi.org/10.1007/s42773-022-00131-8.
- Kataria N, Chauhan AK, Garg VK, Kumar P. Sequestration of heavy metals from contaminated water using magnetic carbon nanocomposites. J Hazard Mater Adv. 2022;6:100066.
- Oyekanmi AA, Katibi KK, Omar RC, Ahmad A, Elbidi M, Alshammari MB, et al. A novel oil palm frond magnetic biochar for the efficient adsorption of crystal violet and sunset yellow dyes from aqueous solution: synthesis, kinetics, isotherm, mechanism and reusability studies. Appl Water Sci. 2024. https://doi.org/10.1007/s13201-023-02060-8.
- Arora B, Dhillayan D, Yadav S, Ghotekar S, Mukherjee S, Mohan C, et al. Bio-fabrication of acid-treated and magnetic biochar from rice straw for tetracycline removal from water: response surface methodology study, reusability, and modelling. Biomass Convers Biorefinery. 2025. https://doi. org/10.1007/s13399-024-06438-9.
- Trinh BS, Le PTK, Werner D, Phuong NH, Luu TL. Rice husk biochars modified with magnetized iron oxides and nano zero valent iron for decolorization of dyeing wastewater. Processes. 2019;7:660.
- Belcaid A, Beakou BH, Bouhsina S, Anouar A. Biosorption of cobalt and chromium from wastewater using manganese dioxide and iron oxide nanoparticles loaded on cellulose-based biochar: modeling and optimization with machine learning (artificial neural network). Int J Biol Macromol. 2024;282:136855.
- Mosaffa E, Ramsheh NA, Patel D, Oroujzadeh M, Banerjee A. Textile industrial wastewater treatment using eco-friendly Kigelia fibrous biochar: column and batch approaches. Process Saf Environ Prot. 2025;194:555–71.
- Han C, Chen F, Lian C, Liang R, Liang W, Chen M, et al. Development of preparation method and application of biochar. Int J Emerg Technol Adv Eng. 2020;10:6–13.
- Li Y, Kumar Awasthi M, Sindhu R, Binod P, Zhang Z, Taherzadeh MJ. Biochar preparation and evaluation of its effect in composting mechanism: a review. Bioresour Technol. 2023;384:129329.
- Ravindiran G, Sundaram H, Rajendran EM, Ramasamy S, Nabil A-Z, Ahmed B. Removal of azo dyes from synthetic wastewater using biochar derived from sewage sludge to prevent groundwater contamination. Urban Clim. 2023;49:101502.
- Zhao Z, Lin L, Zhang J, Xu B, Ma Y, Li J. A green approach to enhance the adsorption capacity: synthesis of bamboo-based adsorbent by biological pretreatment. Ind Crops Prod. 2024;213:118388.
- Srivatsav P, Bhargav BS, Shanmugasundaram V, Arun J, Gopinath KP, Bhatnagar A. Biochar as an eco-friendly and economical adsorbent for the removal of colorants (Dyes) from aqueous environment: a review. Water. 2020;12:3561.
- Eleryan A, Hassaan MA, Aigbe UO, Ukhurebor KE, Onyancha RB, El-Nemr MA, et al. Kinetic and isotherm studies of Acid Orange 7 dye absorption using sulphonated mandarin biochar treated with TETA. Biomass Convers Biorefinery. 2024;14:10599–610.
- Santhosh C, Daneshvar E, Tripathi KM, Baltrénas P, Kim TY, Baltrénaité E, et al. Synthesis and characterization of magnetic biochar adsorbents for the removal of Cr(VI) and Acid orange 7 dye from aqueous solution. Environ Sci Pollut Res. 2020;27:32874–87.
- Zhang P, O'Connor D, Wang Y, Jiang L, Xia T, Wang L, et al. A green biochar/iron oxide composite for methylene blue removal. J Hazard Mater. 2020;384:121286.
- Roy H, Islam MS, Arifin MT, Firoz SH. Chitosan-ZnO decorated Moringa oleifera seed biochar for sequestration of methylene blue: Isotherms, kinetics, and response surface analysis. Environ Nanotechnol, Monit Manag. 2022;18:100752.

- 42. Shi Q, Wang Y, Zhang X, Shen B, Wang F, Zhang Y. Hierarchically porous biochar synthesized with CaCO3 template for efficient Hg0 adsorption from flue gas. Fuel Process Technol. 2020;199:106247.
- Ivanisenko Y, Sauvage X, Mazilkin A, Kilmametov A, Beach JA, Straumal BB. Bulk nanocrystalline ferrite stabilized through grain boundary carbon segregation. Adv Eng Mater. 2018. https://doi.org/10.1002/adem.20180 0443.
- 44. Manikandan A, Yogasundari M, Thanrasu K, Dinesh A, Raja KK, Slimani Y, et al. Structural, morphological and optical properties of multifunctional magnetic-luminescent ZnO@Fe3O4 nanocomposite. Phys E Low-Dimensional Syst Nanostructures. 2020;124:114291.
- Wang S, Gao B, Zimmerman AR, Li Y, Ma L, Harris WG, et al. Removal of arsenic by magnetic biochar prepared from pinewood and natural hematite. Bioresour Technol. 2015;175:391–5.
- Lü T, Ma R, Ke K, Zhang D, Qi D, Zhao H. Synthesis of gallic acid functionalized magnetic hydrogel beads for enhanced synergistic reduction and adsorption of aqueous chromium. Chem Eng J. 2021;408:127327.
- Li H, Dong X, da Silva EB, de Oliveira LM, Chen Y, Ma LQ. Mechanisms of metal sorption by biochars: biochar characteristics and modifications. Chemosphere. 2017;178:466–78.
- Omar H, El-Gendy A, Al-Ahmary K. Bioremoval of toxic dye by using different marine macroalgae. Turk J Botany. 2018;42:15–27.
- Foroutan R, Peighambardoust SJ, Esvandi Z, Khatooni H, Ramavandi B. Evaluation of two cationic dyes removal from aqueous environments using CNT/MgO/CuFe2O4magnetic composite powder: a comparative study. J Environ Chem Eng. 2021;9:104752.
- 50. Li K, Yan J, Zhou Y, Li B, Li X. β-cyclodextrin and magnetic graphene oxide modified porous composite hydrogel as a superabsorbent for adsorption cationic dyes: Adsorption performance, adsorption mechanism and hydrogel column process investigates. J Mol Liq. 2021;335:116291.
- Zbair M, Anfar Z, Ait Ahsaine H, El Alem N, Ezahri M. Acridine orange adsorption by zinc oxide/almond shell activated carbon composite: operational factors, mechanism and performance optimization using central composite design and surface modeling. J Environ Manage. 2018;206:383–97.
- Yang X, Li F, Xia M, Luo F, Jiang Y. Investigation on the micro-structure and adsorption capacity of cellulosic biomass carbon based montmorillonite composite. Microporous Mesoporous Mater. 2018;256:18–24.
- Guo Y, Zeng Z, Liu Y, Huang Z, Cui Y, Yang J. One-pot synthesis of sulfur doped activated carbon as a superior metal-free catalyst for the adsorption and catalytic oxidation of aqueous organics. J Mater Chem A. 2018;6:4055–67.
- Tian W, Zhang H, Sun H, Tadé MO, Wang S. One-step synthesis of flourderived functional nanocarbons with hierarchical pores for versatile environmental applications. Chem Eng J. 2018;347:432–9.
- Wang G, Chen S, Quan X, Yu H, Zhang Y. Enhanced activation of peroxymonosulfate by nitrogen doped porous carbon for effective removal of organic pollutants. Carbon N Y. 2017;115:730–9.
- Hameed BH, Din ATM, Ahmad AL. Adsorption of methylene blue onto bamboo-based activated carbon: kinetics and equilibrium studies. J Hazard Mater. 2007;141:819–25.
- Rose PK, Poonia V, Kumar R, Kataria N, Sharma P, Lamba J, et al. Congo red dye removal using modified banana leaves: adsorption equilibrium, kinetics, and reusability analysis. Groundw Sustain Dev. 2023;23:101005.
- Almehizia AA, Al-Omar MA, Naglah AM, Alkahtani HM, Obaidullah AJ, Bhat MA. Synthesis of low-cost, bio-based novel adsorbent material using charge-transfer interaction for water treatment from several pollutants: waste to worth. Crystals. 2023;13:619.
- Li H, Han K, Shang J, Cai W, Pan M, Xu D, et al. Comparison of adsorption capacity and removal efficiency of strontium by six typical adsorption materials. Sustain. 2022;14:7723.
- Zheng L, Gao Y, Du J, Zhang W, Huang Y, Wang L, et al. A novel, recyclable magnetic biochar modified by chitosan-EDTA for the effective removal of Pb(ii) from aqueous solution. RSC Adv. 2020;10:40196–205.
- Liang M, Ding Y, Zhang Q, Wang D, Li H, Lu L. Removal of aqueous Cr(VI) by magnetic biochar derived from bagasse. Sci Rep. 2020. https://doi.org/ 10.1038/s41598-020-78142-3.
- 62. Budnyak TM, Błachnio M, Slabon A, Jaworski A, Tertykh VA, Deryło-Marczewska A, et al. Chitosan deposited onto fumed silica surface as sustainable hybrid biosorbent for acid orange 8 dye capture: effect of

temperature in adsorption equilibrium and kinetics. J Phys Chem C. 2020;124:15312–23.

- Zhang Y, Hu A, Xiong P, Zhang H, Liu Z. Experimental study of temperature effect on methane adsorption dynamic and isotherm. Energies. 2022;15:5047.
- Horsfall M, Spiff Al. Effects of temperature on the sorption of Pb2+ and Cd 2+ from aqueous solution by Caladium bicolor (Wild Cocoyam) biomass. Electron J Biotechnol. 2005;8:162–9.
- Sun L, Chen D, Wan S, Yu Z. Performance, kinetics, and equilibrium of methylene blue adsorption on biochar derived from eucalyptus saw dust modified with citric, tartaric, and acetic acids. Bioresour Technol. 2015;198:300–8.
- Ahmed MJ, Okoye PU, Hummadi EH, Hameed BH. High-performance porous biochar from the pyrolysis of natural and renewable seaweed (Gelidiella acerosa) and its application for the adsorption of methylene blue. Bioresour Technol. 2019;278:159–64.
- 67. Nethaji S, Sivasamy A. Adsorptive removal of an acid dye by lignocellulosic waste biomass activated carbon: equilibrium and kinetic studies. Chemosphere. 2011;82:1367–72.
- 68. Al-Hazmi GH, Saad HA, Refat MS, Adam AMA. Fe3O4-Carbon-based composite derived from the charge-transfer reaction using waste tea leaves as the carbon precursor for enhanced removing of azocarmine G2, Methyl violet 2B, eosin B, and toluidine blue from aqueous solution. Crystals. 2022;12:1355.
- 69. Wakejo WK, Meshasha BT, Kang JW, Chebude Y. Enhanced ciprofloxacin removal from aqueous solution using a chemically modified biochar derived from bamboo sawdust: adsorption process optimization with response surface methodology. Adsorpt Sci Technol. 2022;2022:2699530.

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