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Magnetically Recoverable Fe–Co–N Functionalized Pomegranate Peel–Derived Pyrolyzed Hydrochar for High-Efficiency Malachite Green Remediation: Integrated Kinetic, Isotherm, and Mechanistic Insights

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Abstract: In this study, waste biomass was strategically valorized to fabricate a magnetically recoverable Fe–Co–N functionalized pyrolyzed hydrochar (Fe–Co–N/HCpp) composite derived from pomegranate peel via a two-stage thermal treatment (300/700 °C), and its performance for Malachite Green (MG) removal from aqueous media was systematically investigated. FT-IR, XRD, SEM and EDX analyses comprehensively elucidated the structural, chemical, and morphological characteristics, confirming effective surface functionalization and successful heteroatom incorporation. The synthesized composite exhibited pronounced magnetic responsiveness, enabling rapid and straightforward adsorption without secondary contamination. Batch adsorption experiments were conducted to assess MG removal efficiency as a function of contact time, adsorbent dosage, initial dye concentration, and solution pH. Equilibrium and kinetic data were rigorously analyzed using established isotherm and rate models to clarify the governing adsorption mechanism. The results revealed that MG removal efficiency significantly increased with increasing pH, attaining a maximum at pH 8, while adsorption capacity increased proportionally with higher initial dye concentrations due to the enhanced mass-transfer driving force. Equilibrium data were best described by the Langmuir isotherm, yielding a maximum monolayer adsorption capacity of 333.40 mg g⁻¹, whereas kinetic analysis demonstrated superior conformity to the pseudo-second-order model. The adsorption system reached equilibrium within 45 min, during which most accessible active sites became saturated, as evidenced by the negligible additional uptake beyond this contact duration. The outstanding adsorption performance of the composite is attributed to synergistic contributions from electrostatic interactions, π – π electron donor–acceptor interactions between aromatic dye molecules and the carbonaceous matrix, surface complexation mechanisms, and coordination interactions involving Fe–Co catalytic centers. Overall, the findings indicate that fruit peel biomass can be effectively transformed into an environmentally sustainable, cost-efficient, and high-performance magnetic adsorbent, offering a promising alternative to commercial adsorbents for advanced wastewater treatment applications.

Keywords: Pomegranate peel, Hydrochar, Fe-Co, Malachite green, Urea

Introduction

Dye-contaminated effluents remain an important environmental concern because they reduce light penetration, suppress photosynthesis, alter oxygen demand and other water-quality parameters, and exert acute or chronic toxicity across aquatic biota. In addition, some dyes and dye-derived products continue to raise mutagenic, carcinogenic, and teratogenic concerns, with potential human exposure occurring through contaminated water,

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irrigated soils, and food webs (Islam et al., 2025). One of the most widely used industrial dyes and organic contaminants that can be discovered in water is malachite green (MG) dye. MG is employed as a food colorant and dyeing agent in the food and textile sectors (Oladoye et al., 2023). MG remains a high-priority model contaminant because it is a cationic dye with persistent use histories in textile and related industrial streams, and because recent resarchs still describe it as environmentally hazardous, toxic, and genotoxic/mutagenic, with continued concern over contaminated wastewaters (Oladoye et al., 2023; Wang et al., 2022).

Pomegranate peel is a logical precursor since it is an plentiful agro-industrial waste that has already been confirmed in numerous nearby carbon-material studies. Sassi et al. (2023) demonstrated that pomegranate-peel biochar can exhibit a high surface area, mesoporous structure, and effective dye uptake capacity. Similarly, Mussa et al. (2025) reported that pomegranate-peel biochar showed strong MG-specific adsorption, achieving 99.85% removal under optimized batch conditions. Pomegranate peel is a promising precursor for MG-relevant adsorbents.

Typical hidrothermal carbonization (HTC) operates at about 180–250 °C under autogenous pressure sufficient to keep water liquid, commonly reported as roughly 10–50 bar, with residence times from about 2 to 12 hours (Khan et al., 2022). Pomegranate peel may be transformed into hydrochar through microwave-assisted hydrothermal carbonization at 200°C for 1 hour, yielding a porous, amorphous material with quantifiable surface charge characteristics and abundant oxygenated functionality, according to Hessien (2022). Accordingly, converting raw pomegranate peel into hydrochar and subsequently into pyrolyzed hydrochar offers a rational strategy for balancing the oxygen-containing surface functionalities generated during hydrothermal processing with the enhanced aromaticity and pore development typically associated with high-temperature carbonization (Mussa et al., 2025).

Recent studies indicate that magnetic hydrochars and magnetized plant-based adsorbents can enhance contaminant uptake by introducing additional active sites and distinctive interfacial chemistry, while also improving adsorbent handling and reducing downstream separation challenges. Although iron oxides remain the most widely used magnetic components because of their stability and relatively low toxicity, more advanced metal configurations may offer additional benefits. In particular, Fe-containing phases provide magnetic responsiveness and metal-related complexation sites, while Co can act synergistically with Fe in bimetallic carbon systems and promote structural ordering of the carbon matrix. Nitrogen doping provides an additional level of functionality by generating defect-rich, electronically modulated surfaces, influencing pore development, and stabilizing chemically diverse binding sites (Zhang et al., 2023; Hu et al., 2025).

Recent MG adsorption studies indicate that the underlying mechanism is governed by multiple concurrent interactions rather than a single dominant pathway. Recurring contributions include pore filling, electrostatic attraction between cationic MG and deprotonated adsorbent surfaces, π - π stacking between the aromatic structure of MG and carbon domains, hydrogen bonding, hydrophobic interactions, n - π interactions, functional-group complexation, and, in some cases, ion exchange. Magnetic hydrochars further expand this mechanistic understanding by highlighting surface complexation and metal-assisted interfacial interactions as important contributors in magnetic carbon-based adsorbents. The most rigorous mechanistic hypothesis is that Fe-Co-N functionalization may enhance MG uptake through the combined effects of pore architecture, defect-rich N sites, aromatic carbon domains, pH-dependent electrostatic interactions, and metal-associated binding. At the same time, the magnetic phase may improve adsorbent recovery and reuse, rather than functioning merely as a passive additive (Meng et al., 2025).

This research aims to functionalize the hydrochar produced from pomegranate peel waste through hydrothermal carbonization followed by pyrolysis with Fe-Co-N to create a magnetic adsorbent that is highly surface reactive and simply recoverable using an external magnetic field. The efficacy of magnetic Fe-Co-N functionalized nano-composite (Fe-Co-N/HCpp) in removing MG, a harmful and cationic dye, from aqueous environments in terms of adsorption capacity, kinetic behavior depending on contact time, equilibrium isotherms, thermodynamic behavior, and reusability after numerous uses will then be thoroughly assessed.

Materials and Experimentation

Iron(III) chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$), cobalt(II) chloride hexahydrate ($\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$), urea ($\text{CO}(\text{NH}_2)_2$) and KOH were obtained from Merck (Darmstadt, Germany). Malachite Green (MG) dye was obtained from Acros Organics (New Jersey, USA). All chemicals used in the experiments were of analytical purity. The biomass was purchased from a local market in Turkey and used to prepare an adsorbent. A pH700 glass-electrode pH meter

from Apera Instruments, a PSU-10 orbital shaker from Biosan, and an FN 500 oven from Nüve were used to prepare the hydrochar. The concentration of MG was measured using a Shimadzu UV-1700 UV–Vis spectrophotometer. FT-IR spectra were recorded with a Thermo Scientific Nicolet iS20 spectrometer, while XRD analysis was performed using a Europe model X-ray diffractometer. The microstructure of the adsorbent was examined using a scanning electron microscope (SEM, Zeiss Gemini SEM 500).

Preparation of Raw Material and Hydrochar

Pomegranate peels (PP) were manually separated from the fruit, thoroughly washed with deionized water to remove surface impurities, and subsequently dried. The dried biomass was initially pulverized using a laboratory mill and passed through a sieve to ensure particle homogeneity. The resulting powder was repeatedly washed with 0.1 mol L⁻¹ HCl and subsequently rinsed with distilled water before being dried at room temperature for 24 h. Then, the dried biomass and deionized water were charged into a stainless-steel autoclave reactor. Hydrothermal carbonization was carried out at 200 °C for 8 h in a temperature-controlled oven. After cooling to room temperature, the obtained hydrochar sample was washed with 0.1 M HCl and rinsed with ultra-pure water until a neutral pH was reached. Finally, the product was oven-dried, ground in a laboratory grinder, and fractionated using a sieve to obtain a definitive particle size range of 200 µm for subsequent composite synthesis.

Synthesis of Adsorbent

The magnetic Fe–Co–N functionalized hydrochar nano-composite (Fe–Co–N/HCpp) was synthesized using a two-stage thermal method. In the first stage, a mixture of pomegranate-derived hydrochar, urea, FeCl₃·6H₂O, and CoCl₂·6H₂O in an aqueous solution was prepared, and this mixture was thermally treated at 300 °C for 1 h to convert the composite into a metal/nitrogen-doped hydrochar intermediate. In the second stage, the resulting intermediate was pyrolyzed in the presence of KOH at 700 °C for 1 h to successfully obtain a nano-composite structure with high porosity and strong magnetic susceptibility. Finally, the synthesized Fe–Co–N/HCpp was washed with pure water and dried in an oven at 60 °C (Figure 1.).

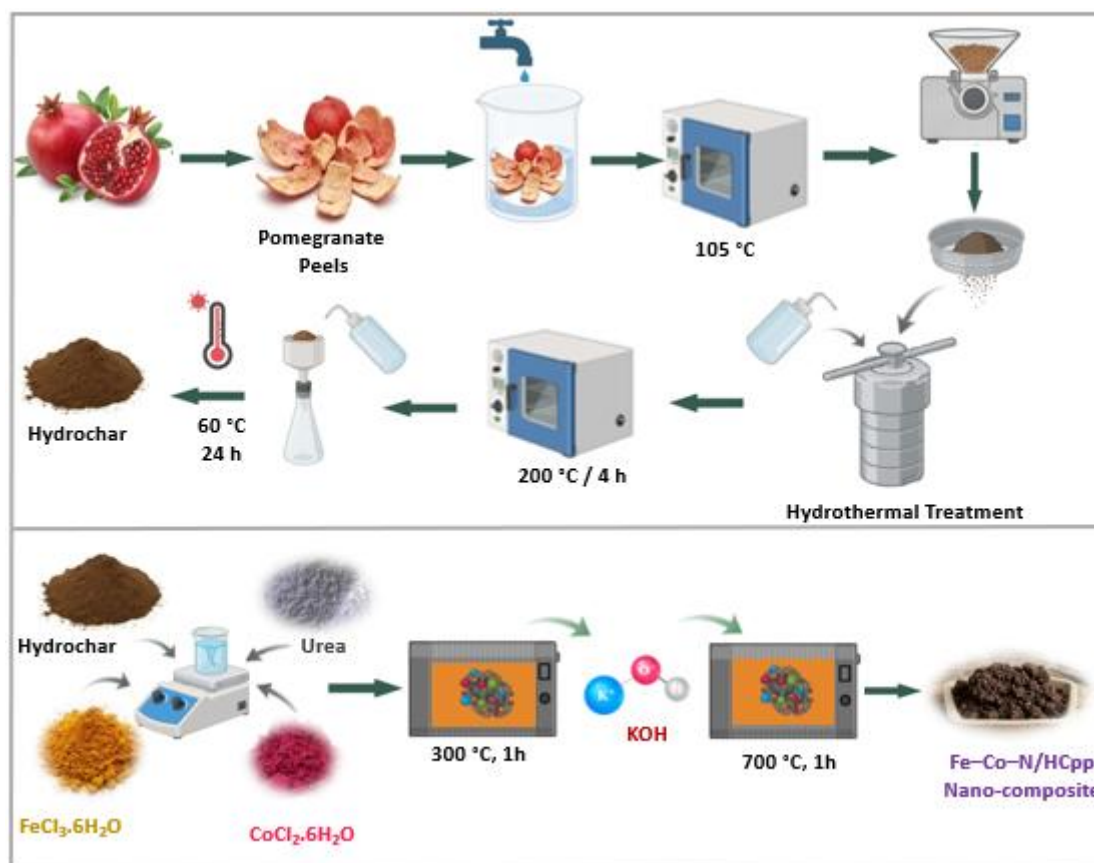


Figure 1. Fe-Co-N/HCpp nano-composite synthesis.

Batch Adsorption Experiments

Batch adsorption experiments for MG removal were systematically carried out in temperature-controlled batch reactors at 25 °C. All analyses were performed in duplicate to ensure the accuracy, reliability, and reproducibility of the experimental findings. Initially, a starting MG concentration of 200 ppm was used; subsequently, various operating parameters were optimized to evaluate their effects on the adsorption capacity and efficiency of Fe–Co–N/HCpp. In this study, the effects of solution pH (3–9), initial MG concentration (100–800 mg L⁻¹), adsorbent amount (0.005–0.04 g), contact time (5–240 min), and reaction temperature (25–45 °C) on MG removal were systematically investigated. After the completion of the adsorption process, the MG concentration remaining in the filtrate was measured at regular intervals using a UV-vis spectrophotometer. The equilibrium adsorption capacity (q_e , mg g⁻¹) and percentage removal efficiency (%) of the composites were determined based on the difference between the initial and final MG concentrations, as expressed in Eqs. (1) and (2).

$$q_e = \frac{(C_0 - C_e)V}{m} \quad (1)$$

$$\text{Removal}(\%) = \frac{(C_0 - C_e)100}{C_0} \quad (2)$$

Results and Discussion

Structural and Morphological Characterization of the Fe–Co–N/HCpp

The surface functional groups and crystal structure of the produced Fe–Co–N/HCpp nano-composite were determined by FTIR and XRD analyses, respectively. Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) analyses were used to examine the surface morphology and elemental composition of the nano-composite. Furthermore, post-adsorption FTIR analyses were performed to confirm MG adsorption on the Fe–Co–N/HCpp surface and elucidate the adsorption by examining structural changes.

FT-IR Spectrum of Composite

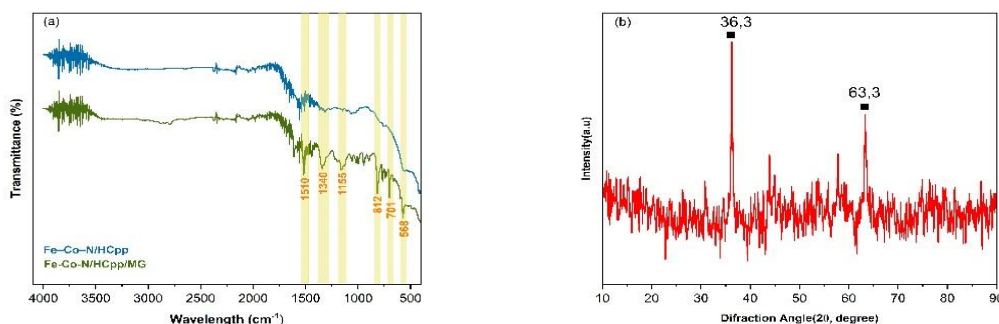


Figure 2. FT-IR diagram of Fe–Co–N/HCpp (a), XRD diagram of Fe–Co–N/HCpp (b).

Figure 2a. shows the FTIR spectra of the Fe–Co–N/HCpp composite before and after MG adsorption. The broad band observed at around 3200–3500 cm⁻¹ in the Fe–Co–N/HCpp composite is attributed to the stretching vibrations of surface hydroxyl (–OH) and amino (–NH) groups. After MG adsorption, the broadening of this band and the increase in its intensity indicate that hydroxyl- and nitrogen-containing functional groups participate in the adsorption mechanism through hydrogen bonding and electrostatic interactions. It has also been previously reported that these groups in hydrocarbon-based adsorbents can form hydrogen bonds and electrostatic interactions with cationic dyes (Hammud et al. 2023).

Distinct changes were observed in the 1700–1500 cm⁻¹ region after adsorption. The increase in the intensity of the peak located at approximately 1510 cm⁻¹ may be associated with aromatic C=C and/or C=N vibrations (Kohzadi et al., 2023). Since MG has a conjugated aromatic structure, this increase indicates that MG molecules are bound to the aromatic carbon structure of Fe–Co–N/HCpp through π – π interactions. Similarly, the increase in the intensity of the C–N stretching vibrations observed at 1340 cm⁻¹ indicates that the nitrogen-containing

functional groups introduced onto the Fe–Co–N/HCpp surface through urea modification interact with the dimethylamino groups in the MG molecule. This is consistent with the study reporting that nitrogen-doped carbon-based adsorbents exhibit higher adsorption performance toward MG molecules (Sun et al. 2015).

A distinct increase in intensity and a change in band shape were observed after adsorption in the band located at approximately 1155 cm^{-1} , which corresponds to C–O, C–O–C, and C–N stretching vibrations (Han et al. 2017). This change indicates that the oxygen-containing functional groups on the Fe–Co–N/HCpp surface directly interact with MG molecules (Zhang et al. 2017). In addition, the appearance of numerous new vibration bands and the formation of more distinct peaks in the spectrum after adsorption, particularly in the fingerprint region between 1500 and 600 cm^{-1} , confirm the presence of characteristic vibrations belonging to MG on the nano-composite surface (Coates, 2000; Zhang et al. 2017). The changes in this region are considered one of the important indicators of the highly effective interaction between Fe–Co–N/HCpp and MG.

The bands observed at approximately 812 and 701 cm^{-1} became more pronounced after adsorption, which can be associated with out-of-plane C–H bending vibrations of aromatic rings. The changes in these bands support the presence of characteristic aromatic vibrations of MG molecules on the composite surface and indicate that the dye molecules interact with the surface functional groups after adsorption (Ojediran et al. 2021). The change in the intensity of the Fe–O and Co–O vibration band observed at around 568 cm^{-1} indicates that Fe and Co active centers play a role in the adsorption mechanism (Oguntuyi et al. 2026; Tabassum et al. 2024). This change suggests the possible occurrence of coordination interactions between MG molecules containing electron-rich amino groups and the metal active centers. Similar FTIR changes have been reported in MG adsorption studies conducted with Fe-based magnetic adsorbents (Parlayıcı et al. 2026; Sun et al. 2015). Overall, the FTIR results indicate that the adsorption of MG onto the Fe–Co–N/HCpp nano-composite occurs through the combined effects of hydrogen bonding, electrostatic attraction, π – π interactions, coordination with Fe/Co centers, and pore-filling mechanisms.

XRD Spectrum of Composite

Examination of the XRD pattern of the Fe–Co–N/HCpp reveals prominent diffraction peaks at angles of $2\theta = 36.2^\circ$ and 63.3° in Figure 2b. These peaks indicate that Fe and Co species form crystalline structures on the carbon matrix during the pyrolysis process (Aboul-Enein et al. 2023). Along with the extensive amorphous structure derived from carbon, these observed diffraction peaks confirm the successful loading of metal species onto the hydrochar surface and their incorporation into the nano-composite structure. The results demonstrate the successful formation of Fe and Co-containing active centers on the carbon structure. Similarly, it has been reported in the literature that metal-containing crystalline phases form in Fe–Co modified biocarbon systems while the carbon structure largely retains its amorphous character (Rehman et al. 2024; Abdelrahman et al. 2024).

SEM–EDX Analysis of the Composite

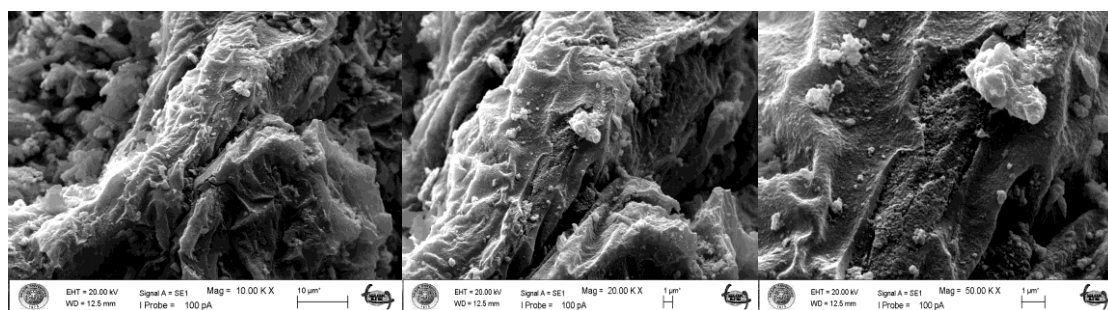


Figure 3. SEM image of Fe-Co-N/HCpp.

SEM images of the Fe–Co–N/HCpp reveal an irregular, heterogeneous, and distinctly rough surface morphology. Particle clusters of varying sizes are observed on the layered structure formed as a result of hydrothermal carbonization and subsequent modification processes (Figure 3.). The voids and irregular structures on the surface create additional active sites for adsorption, exhibiting properties that may facilitate the retention of dye molecules. Furthermore, the bright and dense regions observed on the surface are thought to indicate the successful dispersion of Fe and Co-containing metal species onto the hydrochar matrix. This morphological structure supports the finding of a high adsorption capacity.

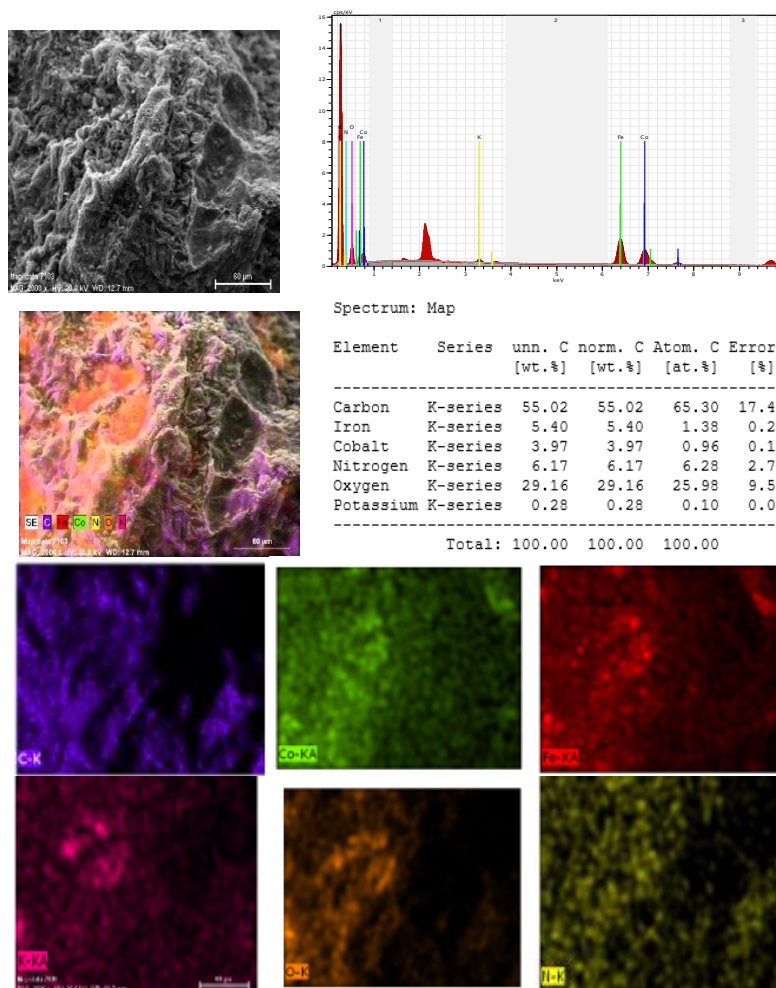


Figure 4. SEM-EDX mapping images of the Fe-Co-N/HCpp composite.

EDX analysis (Figure 4.) shows that the nano-composite is primarily composed of carbon (55.02%), oxygen (29.16%), nitrogen (6.17%), iron (5.40%) and cobalt (3.97%) elements. The high carbon content confirms the formation of a carbon-rich structure as a result of hydrothermal carbonization. The presence of oxygen indicates the presence of oxygen-containing functional groups such as hydroxyl, carbonyl, and carboxyl in the structure. The presence of nitrogen in the composite shows that nitrogen was successfully incorporated into the carbon structure as a result of urea modification. The detection of Fe and Co signals in the EDX spectrum confirms that Fe and Co species were successfully loaded onto the hydrochar surface. The presence of Fe not only confers magnetic properties to the composite but also affective during adsorption, improving dye removal performance. The low amount of K (0.28%) detected may originate from activation processes.

Results of the Batch Adsorption Experiments

Effect of pH

To investigate the effect of pH on MG adsorption, the initial pH values of the dye solutions were adjusted to a range of 3.0–9.0. The pH of the solution affects the functional groups on the Fe-Co-N/HCpp surface, causing changes in surface charge properties and thus significantly influencing adsorption performance. For the cationic dye MG, the decrease in adsorption capacity at low pH values is mainly associated with the abundance of H^+ ions, which compete with MG molecules for active adsorption sites on the Fe-Co-N/HCpp surface. In addition, protonation of the surface functional groups may generate a positively charged surface, thereby weakening the electrostatic attraction toward MG cations. As the pH increases, deprotonation of carboxyl and hydroxyl groups on the Fe-Co-N/HCpp surface enhances its negative surface charge, thereby strengthening the electrostatic attraction between the positively charged MG molecules and the hydrochar surface. This allows the dye molecules to bind more effectively to the Fe-Co-N/HCpp surface, contributing to an increase in adsorption capacity. In addition to electrostatic interactions, π - π interactions and hydrogen bonds can also contribute to the adsorption

mechanism. Similar results are supported by studies reporting that hydrochars obtained by hydrothermal carbonization exhibit pH-dependent increased adsorption performance in cationic dye removal (Hammud et al., 2023; Kohzadi et al., 2023). The presence of Fe in the Fe-Co-N/HCpp may contribute to MG adsorption by introducing additional surface-active sites, such as Fe-O, Fe-OH, or Fe-N functionalities, which can promote electrostatic interactions, surface complexation, and Lewis acid-base interactions with MG molecules, thereby enhancing the overall dye removal performance. Changes in the adsorption capacity of MG under different pH conditions are shown in Figure 5.

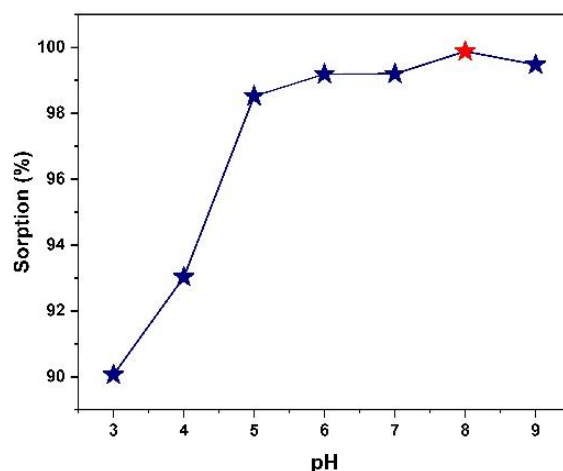


Figure 5. The effect of pH on MG adsorption.

Effect of Fe-Co-N/HCpp Amount

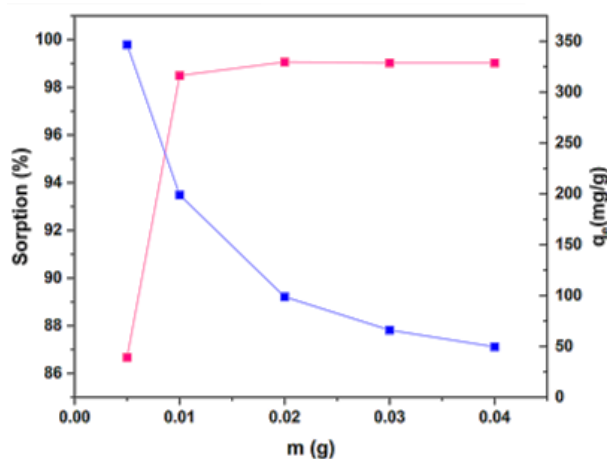


Figure 6. Effect of Fe-Co-N/HCpp composite amount.

The influence of adsorbent amount on MG adsorption was evaluated by varying the adsorbent mass from 0.005, 0.01, 0.02, 0.03, and 0.04 g. Adsorption performances obtained at different adsorbent amounts are presented in Figure 6. In general, a significant increase in MG removal efficiency was observed as the number of available active sites and the total surface area for adsorption increased with increasing Fe-Co-N/HCpp amount. Adsorption capacity increased up to an adsorbent mass of 0.01 g, and this value was determined as the optimum adsorbent amount. However, it was observed that the increase in removal efficiency remained quite limited at adsorbent doses above 0.01 g. Therefore, 0.01 g was used as the optimum adsorbent mass in subsequent experiments. This situation can be explained by the fact that the active sites approach saturation as a result of the large adsorption of MG molecules in the solution. Similar results have been reported in dye removal studies performed with hydrochar-based adsorbents, where no significant change in adsorption performance was observed after a certain optimum adsorbent dose (Qiao et al., 2024; Supee and Zaini, 2023).

Effect of Fe-Co-N/HCpp Contact Time

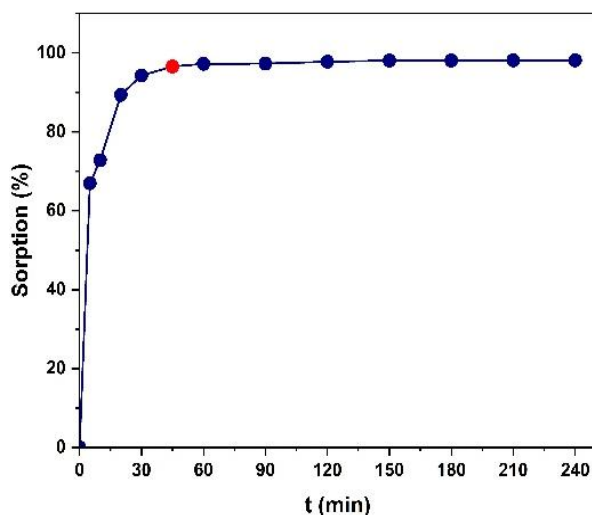


Figure 7. Effect of Fe-Co-N/HCpp contact time.

The effect of contact time on MG adsorption was investigated at intervals of 5, 10, 20, 30, 45, 60, 90, 120, 150, 180, 210, and 240 minutes. As shown in Figure 7, the adsorption capacity increased with increasing contact time, indicating that MG uptake by the Fe-Co-N/HCpp composite was time-dependent. The rapid increase in adsorption capacity during the initial stage can be attributed to the availability of numerous vacant active sites on the adsorbent surface, which facilitated the fast uptake of MG molecules within the first 5 min. As the contact time increased, the adsorption rate slowed down due to the gradual filling of the active sites and the decrease in MG concentration in the solution. In this study, it was observed that the change in adsorption capacity remained quite limited after a certain contact time, indicating that the system reached adsorption equilibrium. Therefore, 45 minutes was chosen as the optimum contact time in subsequent experiments. Similarly, studies conducted with hydrochar-based adsorbents have reported rapid dye retention in the initial stages of adsorption (Kohzadi et al., 2023).

Effect of Dye Initial Concentration

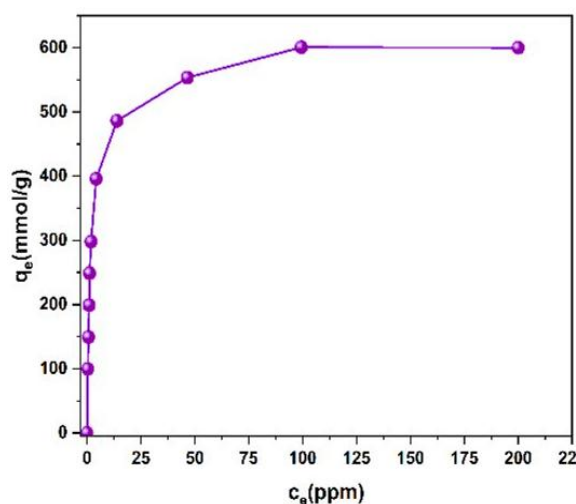


Figure 8. Effect of MG concentration.

To investigate the effect of initial dye concentration on adsorption performance, experiments were conducted using MG solutions prepared in the concentration range of 100–800 mg L⁻¹. As the initial concentration increased, the number of MG molecules in the solution increased, increasing the concentration gradient between the adsorbent and adsorbate and contributing to the improvement of the driving force required for mass transfer. Therefore, it was observed that the adsorption capacity increased with increasing initial concentration (Figure 8.). Similarly, studies on hydrothermal carbonization-derived carbon materials have also reported that high initial dye concentrations increase the adsorption capacity (Parlayıcı and Pehlivan, 2026). Langmuir, Freundlich, and Temkin isotherm models were applied to explain the adsorption mechanism of the equilibrium data. Analysis of the

obtained correlation coefficients revealed that the Langmuir model represented the experimental data more successfully than the Temkin and Freundlich models (Table 1.).

Table 1. Adsorption isotherm parameters for removal of MG dye.

Model	Equation	Parameters for dye			
Langmuir	$\frac{C_e}{q_e} = \frac{C_e}{A_s} + \frac{1}{K_b A_s}$	A_s	K_b	R^2	R_L
		333.40	1.77	0.987	0.003
Freundlich	$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e$	K_f	n	R^2	
		198.98	3.86	0.854	
Temkin	$q_e = B_T \ln A_T + B_T \ln C_e$	B_T	A_T	R^2	
		83.13	14.09	0.961	

This indicates that MG adsorption occurs predominantly via a monolayer adsorption mechanism in active sites with homogeneous energy on the Fe–Co–N/HCpp surface. The maximum adsorption capacity ($q_{\max}$) calculated from the Langmuir model was found to be 333.40 mg g⁻¹, demonstrating the effectiveness of the adsorbent in MG removal. Furthermore, the calculation of the Langmuir separation factor (R_L) as 0.003 shows that the adsorption process is quite suitable and feasible. Similarly, it has been reported in various studies that the Langmuir model is dominant in cationic dye adsorption and that the adsorption exhibits a monolayer character (Pehlivan and Parlayıcı, 2024; Sorour et al., 2024).

Effect of Kinetic Studies

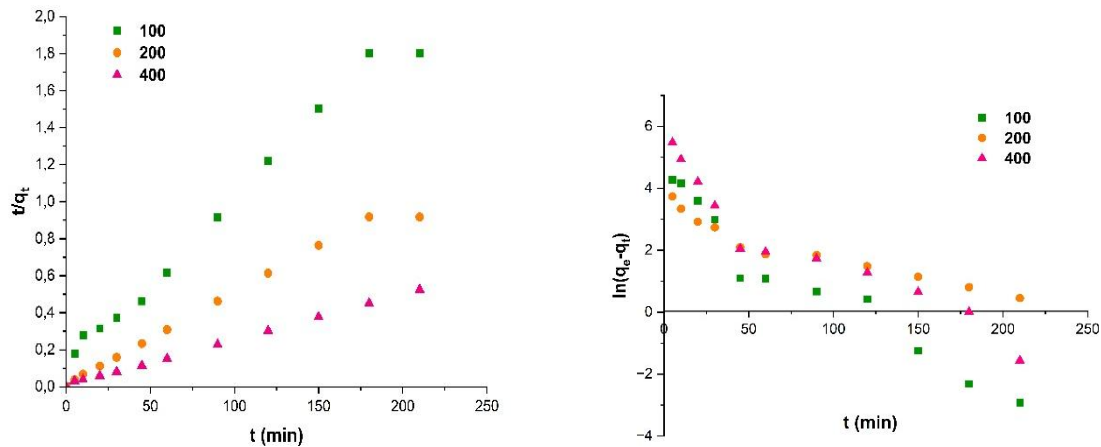


Figure 9. Kinetics for MG dye adsorption onto Fe-Co-N/HCpp nano-composite.

Adsorption kinetic curves for MG removal with Fe-Co-N/HCpp are shown in Figure 9. Table 2 presents the parameter results of both the pseudo-first-order and pseudo-second-order kinetic models used to evaluate the adsorption data. The pseudo-second-order model showed a better fit to the experimental results compared to the pseudo-first-order model. This is confirmed by the higher correlation coefficient (R^2) obtained for the pseudo-second-order model. In conclusion, the adsorption process is more accurately explained by so-called second-order kinetics, suggesting that chemical adsorption and electrostatic attraction are highly likely to be the dominant mechanisms (Yuan & Lu, 2024). Electrostatic attraction, π - π interactions, and hydrogen bonds occurring between carboxyl, hydroxyl, and other oxygen-containing functional groups on the hydrochar surface and MG molecules are thought to contribute to the adsorption mechanism (Qiao et al., 2024). These results reveal that the synthesized hydrochar develops strong surface interactions with MG molecules and that the adsorption process cannot be explained solely by physical diffusion.

Table 2. Comparison of the pseudo-first-order, pseudo-second-order adsorption rate constants and calculated and experimental q_e values obtained at different initial dye concentrations.

C_0 (ppm)	q_e Exp.	Pseudo-first-order			Pseudo-second-order		
		k_1	q_e	R^2	k_2	q_e	R^2
100	99.95	0.0011	50.30	0.947	0.0007	113.64	0.985
200	196.30	0.0014	60.34	0.911	0.0010	212.77	0.989
400	398.45	0.0013	143.70	0.901	0.0007	416.67	0.992

Thermodynamic Analysis of the Adsorption Process

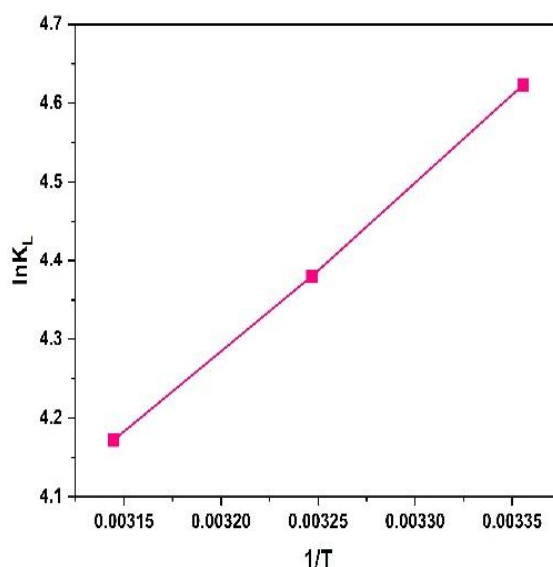


Figure 10. The impact of temperature on dye adsorption.

The effect of temperature on the adsorption of MG onto Fe-Co-N/HCpp was investigated (Figure 10.), and the obtained thermodynamic parameters are presented in Table 3. The negative Gibbs free energy change (ΔG°) values indicate that the adsorption process occurs spontaneously and is thermodynamically favorable (Akiode et al., 2023). Furthermore, the fact that the ΔG° values become less negative with increasing temperature reveals that adsorption is more advantageous at lower temperatures. The negative enthalpy (ΔH°) indicates that the adsorption of MG onto the Fe-Co-N/HCpp surface is exothermic. However, the negative entropy (ΔS°) values suggest that the system disorder at the adsorbent-adsorbate interface decreases during adsorption, and the MG molecules form a more ordered structure on the adsorbent surface. When these results are considered together, it is understood that MG adsorption is a spontaneous, exothermic process that is more favorable at low temperatures.

Table 3. Thermodynamic parameters for adsorption of MG.

$\Delta S^\circ (\text{J K}^{-1} \text{mol}^{-1})$	$\Delta H^\circ (\text{J mol}^{-1})$	$\Delta G^\circ (\text{J mol}^{-1})$			
		T=298.15K	T=308.15K	T=318.15K	R ²
-21.3	-17793.6	-11454.5	-11216.0	-11029.7	0.999

Regeneration and Reusability Performance of the Fe-Co-N/HCpp

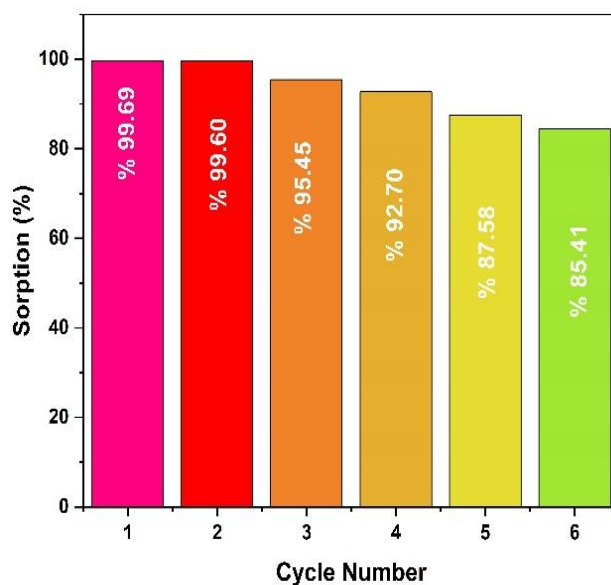


Figure 11. Desorption efficiency after recycle use.

Six consecutive adsorption-desorption cycles were performed to evaluate the reusability of the adsorbent. The results showed that the adsorption efficiency, which was 99.69% in the first cycle, decreased to 85.41% at the end of the sixth cycle (Figure 11.). This observed decrease can be attributed to the irreversible occupation of some of the active adsorption sites by MG molecules and small adsorbent losses occurring during the regeneration process. Nevertheless, the maintenance of high removal efficiency after six cycles reveals that the synthesized Fe-Co-N/HCpp exhibits good reusability and structural stability. These results demonstrate that the adsorbent has the potential to be used as an economical and sustainable alternative in wastewater treatment.

Conclusion

This study investigated the adsorption performance of an Fe-Co-N/HCpp nano-composite for the removal of MG from an aqueous medium. Utilizing plant waste through appropriate methods offers a significant approach in terms of both reducing environmental pollution and developing adsorbents with high economic value. In this context, the synthesized Fe-Co-N/HCpp nano-composite was characterized using FTIR, SEM, EDX, and XRD analyses, and the results confirmed the successful synthesis of the composite. The optimum conditions for adsorption studies were determined as: solution pH 8.0, temperature 25 °C, adsorbent amount 0.01 g, and contact time 45 minutes. The effect of initial MG concentration on adsorption was investigated, and equilibrium data were evaluated using different isotherm models. The results showed that the adsorption process best fit the Langmuir isotherm model. The maximum adsorption capacity calculated from the Langmuir model was found to be 333.40 mg g⁻¹, demonstrating that the synthesized composite exhibits high performance in MG removal. Kinetic studies, considering the obtained R², determined that the adsorption behavior better fits the pseudo-second-order kinetic model. This indicates that the adsorption process is largely controlled by surface interactions and chemical adsorption mechanisms. It was assessed that MG adsorption occurs through the combined effect of multiple mechanisms such as electrostatic attraction, π - π interactions, hydrogen bonds, and hydrophobic interactions. Temperature studies showed that the adsorption capacity decreased with increasing temperature. Thermodynamic parameters revealed that the adsorption process is exothermic and spontaneous. Furthermore, the iron content imparts magnetic properties to the composite, enabling facile separation of the adsorbent from the solution using a magnet after adsorption, without requiring filtration or centrifugation. In conclusion, the synthesized Fe-Co-N/HCpp composite has been evaluated as a promising adsorbent for the removal of cationic dyes from textile and other industrial wastewaters due to its high adsorption capacity, suitable kinetic and thermodynamic properties, easily recoverable structure, and the use of a sustainable raw material source. The development of such adsorbents derived from plant waste can make significant contributions to the dissemination of environmentally friendly and economical wastewater treatment technologies.

Scientific Ethics Declaration

* The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

Conflict of Interest

* The authors declare that they have no conflicts of interest.

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