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ORIGINAL RESEARCH ARTICLE

Green synthesis of ricinus communis leaf adsorbents for methylene blue dye removal: Adsorption, regeneration, and reuse

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ABSTRACT

This study evaluated the potential of using Ricinus communis Leaves, which are often regarded as agricultural waste, as a source of activated carbon for removing organic dyes like methylene blue (MB) from water solutions. This approach presents a practical and cost-effective method for converting a waste product into a valuable resource. The castor plant was activated using hydrochloric acid to enhance its surface area and efficiency. Various process parameters were assessed, including the concentration of the initial dye, the weight of the activated carbon, and the pH of the solution. The experimental results showed a significant increase in dye removal efficiency from 68.22% to 97.23% as the adsorbent dosage increased from 0.01 g to 0.1 g. This improvement is attributed to the increased number of available active sites on the activated carbon surface, which enhances the interaction between the adsorbent and the dye molecules. Additionally, the results showed that the removal rate (%) of MB dye decreased significantly as the initial concentration of the dye increased, while the removal rate improved with a greater amount of activated carbon. This study suggests that biomass derived from activated carbon is a promising, cost-effective alternative for dye adsorption, potentially offering a better solution than conventional activated carbon methods. Overall, this research emphasizes the importance of optimizing surface efficiency for effective dye removal. Furthermore, a comparison between unactivated Ricinus communis leaves and activated carbon was also conducted.

Keywords: Activated carbon; removal; adsorption; agricultural waste; ricinus communis

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1. Introduction

Water pollution caused by industrial liquid waste, agricultural runoff, and untreated sewage is a significant concern due to toxic substances such as dyes, heavy metals, and organic pollutants. Among various water treatment techniques, adsorption has emerged as one of the most efficient, economical, and environmentally friendly methods for removing a wide range of contaminants from water [1-3]. Adsorption is a surface-based process where polluted molecules adhere to the surface of a solid. The effectiveness of adsorption depends on several factors, including the surface area, pore size distribution, and the type of functional groups present on the

adsorbent surface. This technology offers several advantages, such as simplicity of design, ease of operation, low cost, and high efficiency, even at low concentrations of pollutants [4-7] .

Common materials used for adsorption include activated carbon, clay minerals, biochar, zeolites, and various natural or modified agricultural wastes. Activated carbon derived from biomass has gained attention for its sustainability, high surface area, and favorable surface chemistry. In addition to adsorption, other treatment methods include membrane filtration, chemical precipitation, ion exchange, and advanced oxidation processes. However, these methods may face drawbacks such as high operational costs, the potential for secondary pollution, or limited effectiveness against certain pollutants(8). In contrast, adsorption provides a promising approach for both small and large-scale water treatment applications, especially when utilizing low-cost and renewable materials. Current research is focused on enhancing adsorption efficiency, promoting the regeneration of adsorbent materials, and developing advanced composite materials designed for specific types of pollution[9-11].

Methylene blue (MB) is a cationic dye commonly used for coloring, microbiology, surgery, and diagnostics Figure 1. Although MB is not highly hazardous, it can still cause various harmful effects. Acute exposure to MB may result in increased heart rate, shock, formation of Heinz bodies, cyanosis, jaundice, quadriplegia, and tissue necrosis in humans. Additionally, MB can cause eye burns, which may lead to permanent injury to the eyes of both humans and animals. Therefore, treating effluents containing this dye is crucial due to its adverse impacts on receiving waters. In general, dyes are poorly biodegradable or resistant to environmental conditions, creating significant challenges in treating wastewater containing these substances [12, 13]. A range of technologies has been employed to remove dye contaminants from sewage, including coagulation, foam flotation, precipitation, ozonation, ion exchange, filtration, solvent extraction, electrolysis, chemical oxidation, membrane technology, liquid-liquid extraction, and adsorption on activated carbon. Among these methods, adsorption has proven to be an efficient and cost-effective process for removing dyes, pigments, and other colorants and controlling biochemical oxygen demand [14-16].

Ricinus communis, commonly known as the castor plant, is a fast-growing, widely available species traditionally cultivated for its oil-rich seeds. Beyond its industrial oil applications, increasing attention has been directed toward the potential of its agricultural waste, particularly its leaves, as a low-cost and eco-friendly precursor for producing activated carbon. The abundant cellulose, hemicellulose, and lignin content in Ricinus communis leaves makes them ideal for thermal and chemical activation to generate porous carbonaceous materials with high surface area and functional groups favorable for adsorption. Activated carbon derived from Ricinus communis leaves has demonstrated significant potential in removing various environmental contaminants from aqueous solutions, including dyes, heavy metals, and organic pollutants [-17 19]. This bio-based adsorbent offers a sustainable alternative to conventional materials due to its renewable nature, low production cost, and minimal environmental impact. Moreover, chemical modification or activation using agents such as phosphoric acid or potassium hydroxide enhances its adsorption properties, making it suitable for application in water purification and wastewater treatment systems. This approach supports waste valorization and pollution control and aligns with green chemistry and circular economy principles by transforming agricultural residues into valuable adsorbent materials for environmental remediation[22-20] .

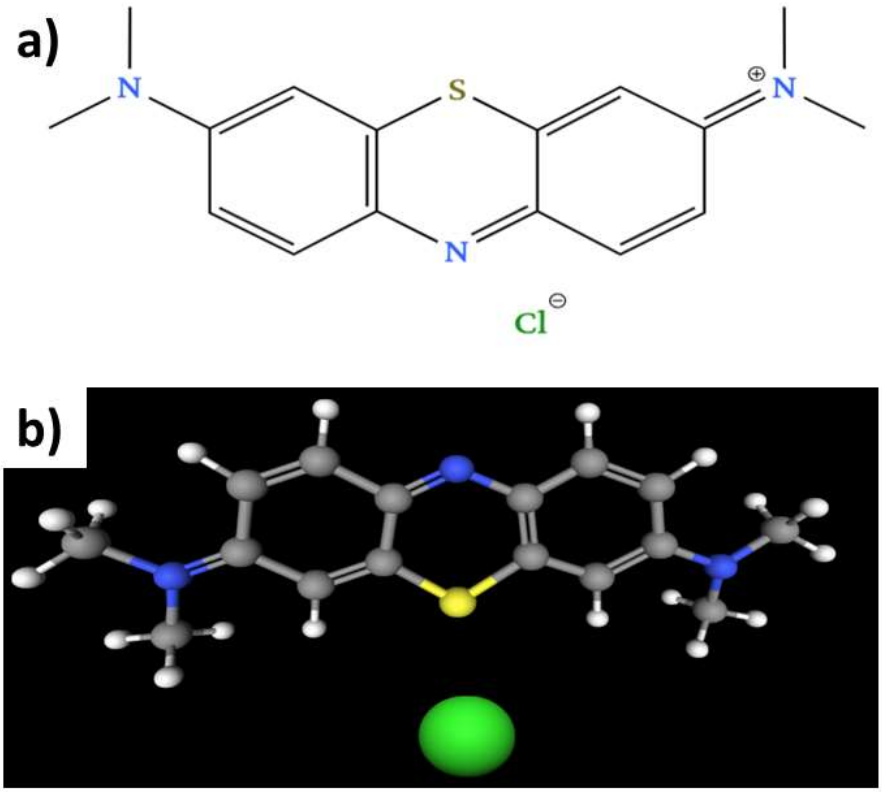

Figure 1. Chemical structure of methylene blue (MB) dye: (a) 2D view, (b) 3D view (created by the authors using MolView software)

1.1. Preparation of ricinus communis-based activated carbon

Fresh Ricinus communis leaves were collected from Iraqi market. The leaves were carefully washed with distilled water to remove impurities and dust. They were then air-dried at room temperature for 48 hours. After drying, the leaves were cut into smaller pieces and further dried in an oven at 80 °C for 24 hours to eliminate any remaining moisture.

Specifically, 10 g of dried Ricinus communis leaves were impregnated with 100 mL of 20% v/v HCl solution (1:10 w/v impregnation ratio). The 20% v/v HCl solution was prepared by diluting concentrated HCl (37%) with deionized water.The impregnation process was performed under constant stirring for two hours to ensure thorough acid penetration into the leaves. Following this step, the saturated biomass was filtered and dried in an oven at 80 °C for another 12 hours. The chemically treated sample was then subjected to pyrolysis in the oven. The pyrolysis was carried out in a muffle furnace. To minimize oxygen exposure, the samples were placed in a covered ceramic crucible during heating. The temperature was increased from room temperature to 300 °C at a heating rate of 10 °C/min and maintained for 2 h under limited-oxygen conditions. After cooling to room temperature, the resultant material was washed multiple times with distilled water until the pH of the filtrate reached approximately 6.6 to 6.8, indicating the removal of any residual acid. Subsequently, the washed material was dried at 105 °C for 6 hours in a hot air oven. Finally, the dried material was processed using a mechanical mill and passed through a 75-micron sieve to obtain a fine powder suitable for further studies. The prepared activated carbon was stored in tightly sealed containers for subsequent characterization and experimental use. For comparison, raw (unactivated) carbon was prepared by subjecting dried Ricinus communis leaves to thermal carbonization at 300 °C for 1 h in a muffle furnace under limited-oxygen conditions (sealed crucible). These conditions were selected to match those applied to the precursor of AC-HCl prior to acid activation.

To ensure clarity and reproducibility, the detailed synthesis conditions used for the preparation of hydrochloric acid–activated carbon from *Ricinus communis* leaves are summarized in **Table 1**. This table

provides a concise overview of the precursor material, activating agent, impregnation ratio, temperature, time, and atmosphere applied during the synthesis process.

Table 1. Summary of synthesis conditions for the preparation of hydrochloric acid–activated carbon from *Ricinus communis* leaves

Precursor	Activating Agent	Impregnation Ratio (w/w)	Temperature (°C)	Time (h)	Atmosphere
Ricinus communis leaves	HCl (20%)	1:2	300	2	Air

Note: *The synthesis conditions were selected based on preliminary optimization experiments. An HCl concentration of 20% was chosen for its cost-effectiveness, safety, and ability to introduce oxygen-containing functional groups. A carbonization temperature of 300 °C and a reaction time of 2 h provided a balance between pore development and material stability. Activation in air was used to ensure process simplicity and reproducibility.*

1.2. Adsorption study

Standard solutions of methylene blue (MB) dye (0.1 g dissolved in 1000 mL distilled water) were prepared, and a series of dilutions was carried out to obtain working solutions in the concentration range of 5–60 mg/L. For each adsorption experiment, 0.05 g of AC-HCl was added to a conical flask containing 100 mL of MB solution at the desired concentration. The flasks were maintained in a thermostatic shaker water bath at a controlled temperature to ensure proper mixing and equilibrium conditions. The adsorption experiments were conducted in a thermostatic shaker bath operated at 150 rpm and 25 ± 1 °C. Samples for kinetic analysis were collected at intervals of 5, 10, 20, 30, 45, and 60 min. Separation of the adsorbent was achieved by centrifugation at 4000 rpm for 10 min. The residual concentration of MB was determined spectrophotometrically (UV–Vis 1700, Shimadzu) at 630 nm, corresponding to the maximum absorbance wavelength of MB. At predetermined time intervals, aliquots were withdrawn and centrifuged to separate the adsorbent. The absorbance of the supernatant was then measured, and the MB concentration in solution was calculated.

$$E\% = \frac{C_o - C_e}{C_o} \times 100 \quad (1)$$

$$Q_e = \frac{(C_o - C_e)V}{m} \quad (2)$$

where C_o (mg/L) is the initial concentration of MB, Ce (mg/L) is the concentration of the MB at equilibrium, V (mL) is the volume of the solution, and m (g) is the mass of the adsorbent.

2. Results and discussion

2.1. Characterization for adsorbent/adsorbate

The surface morphology and microstructure of carbon extracted from castor bean (Ricinus communis) leaves were carefully examined using scanning electron microscopy (SEM) and transmission electron microscopy (TEM) to evaluate the effects of acid activation and dye adsorption. SEM analysis of the raw (unactivated) carbon extracted from castor bean leaves showed a smooth and relatively dense surface with minimal porosity (Figure 2a). The structure appeared compact, indicating limited surface area and few adsorption sites. However, significant changes in surface texture were observed after chemical activation with hydrochloric acid (HCl). The activated carbon exhibited a rougher morphology with numerous cracks, pores, and deformations on its surface, demonstrating the successful development of a porous structure. These newly formed pores increased the surface area and provided more active sites for dye adsorption (as shown in Figure 2b and Figure 2c. After adsorption of methylene blue dye onto acid-activated carbon, scanning electron microscopy (SEM) images revealed additional modifications. The previously visible pores appeared to be fully or partially filled, and the surface was coated with dye molecules. This confirmed that adsorption had occurred and that the activated carbon effectively removed the dye from the aqueous solution [23, 24].

Transmission electron microscopy (TEM) analysis provided insights at the nanoscale. Before dye adsorption, TEM images of the hydrochloric acid-activated carbon showed well-defined nanoparticles, typically in the 100 nm range, and a network of visible micro- and mesopores. These properties support a high surface area, which is critical for adsorption efficiency. Figure 2d. Overall, TEM results demonstrate that hydrochloric acid activation of castor bean leaves improves the material's structural properties, leading to enhanced porosity and surface activity. Moreover, the visual changes after dye adsorption confirm that the modified carbon effectively reacts with and removes methylene blue from aqueous solutions, highlighting its potential as an environmentally friendly adsorbent in wastewater treatment applications[25-27].

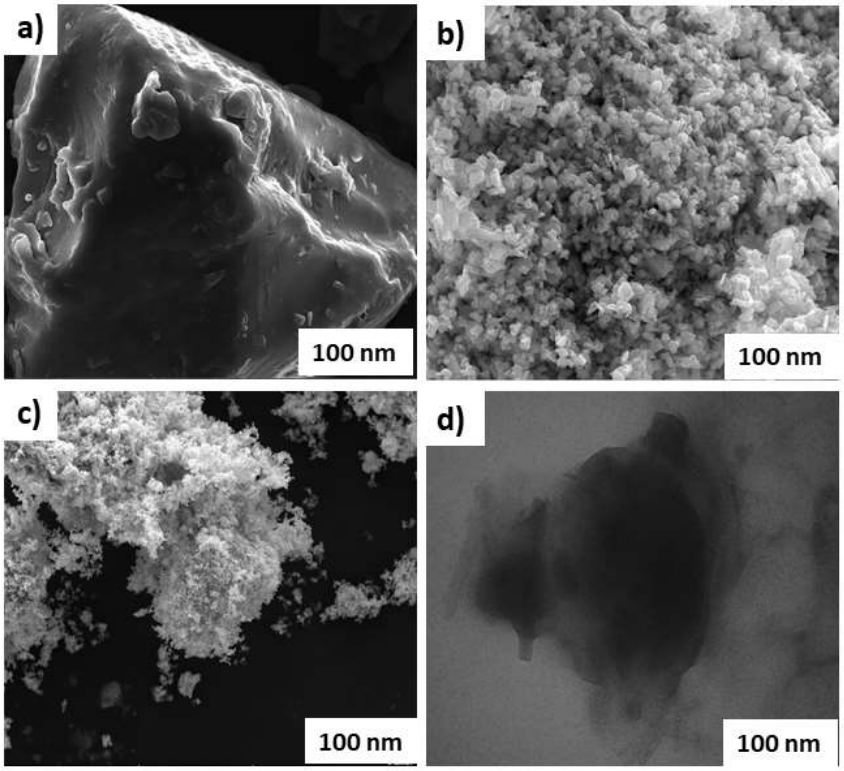

Figure 2. a) Ricinus communis leaves, b) AC-HCl, c) AC-HCl after adsorption FESEM image, and d) TEM image of AC-HCl

Fourier transform infrared (FTIR) spectroscopy was employed to characterize the surface functional groups of the hydrochloric acid–activated carbon derived from Ricinus communis leaves. The spectrum exhibited multiple absorption bands, confirming the incorporation of oxygen-containing groups that play a critical role in adsorption. A broad and intense band centered at ~3400 cm^{-1} corresponds to –OH stretching vibrations of surface hydroxyl groups, which are known to provide hydrogen-bonding sites for dye molecules. The band at ~2900 cm^{-1} is associated with C–H stretching of aliphatic hydrocarbons, indicating the partial preservation of lignocellulosic structures. A strong peak observed at 1700–1720 cm^{-1} is attributed to C=O stretching vibrations from carbonyl and/or carboxylic acid groups, which enhance electrostatic interactions with cationic dyes.[28, 29]. Additionally, the band near 1600 cm^{-1} reflects C=C stretching of aromatic domains, while peaks around 1400 cm^{-1} correspond to C–O stretching of phenolic groups. The signals in the 1100–1000 cm^{-1} region can be ascribed to C–O–C stretching vibrations of ether or ester linkages. The relative intensity of oxygenated functional groups (–OH, C=O, and C–O) suggests a substantial increase in surface polarity due to acid activation, thereby improving the affinity of the adsorbent toward methylene blue. These functional groups act synergistically as active sites, enabling adsorption through hydrogen bonding, electrostatic attraction, and π–π stacking interactions, consistent with the adsorption mechanism proposed in this study (Figure 3). [30].

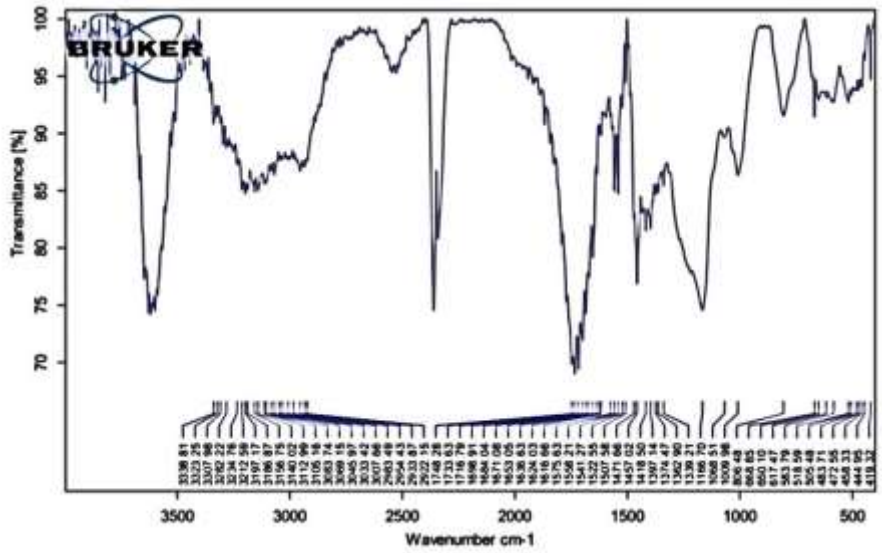

Figure 3. Fourier transform infrared (FTIR) spectroscopy of AC-HCl

X-ray diffraction (XRD) analysis was carried out to evaluate the crystallinity and structural characteristics of the hydrochloric acid–activated carbon derived from *Ricinus communis* leaves. The diffraction pattern revealed a broad hump in the region of 2θ = 22°–26°, which corresponds to the (002) reflection of disordered graphitic carbon. The significant broadening and low intensity of this band indicate that the material is largely **amorphous in nature**, consisting of randomly oriented and poorly stacked graphene layers, which is typical for biomass-derived activated carbons. The absence of sharp crystalline peaks suggests the successful removal of mineral phases and inorganic impurities during acid activation and subsequent washing. This confirms the production of a relatively pure carbonaceous structure. While no precise crystallite size calculations (e.g., Scherrer equation) were performed in this work, the diffuse (002) peak implies the dominance of nanoscale graphitic domains with limited ordering. Importantly, such an amorphous carbon framework is advantageous for adsorption, as it facilitates the development of micropores and mesopores, enhances the surface heterogeneity, and provides a higher density of accessible active sites for interaction with dye molecules. This structural disorder therefore contributes directly to the improved adsorption performance observed in the present study [31, 32].

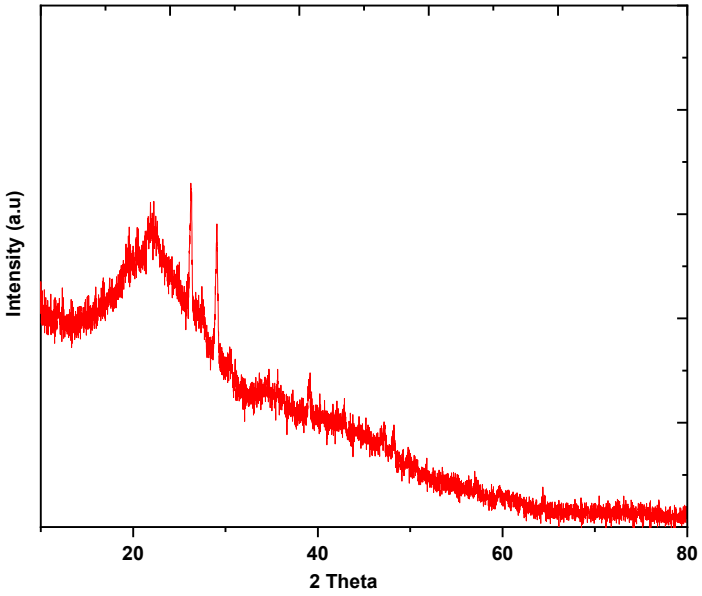

Figure 4. X-ray diffraction (XRD) analysis of AC-HCl

2.2. Adsorbent dosage and its effect on MB dye removal efficiency

The efficiency of an adsorbent in removing contaminants is significantly affected by its quantity (or dosage). To evaluate this, the AC-HCl dosage on the MB dye removal efficiency was studied over a range of 0.01 to 0.1 g, at a constant pH of 7 and an initial MB dye concentration of 50 mg/L, as shown in Figure 5. The experimental results showed a significant increase in dye removal efficiency from 68.77% to 97.99% as the adsorbent dosage increased from 0.01 g to 0.1 g. This improvement is attributed to the increased number of available active sites on the activated carbon surface, which enhances the interaction between the adsorbent and the dye molecules. However, increasing the adsorbent dosage beyond 0.05 g (i.e., at 0.10 g) did not result in a commensurate improvement in removal efficiency. This plateau effect is a well-documented phenomenon in adsorption studies, often attributed to adsorbent surface saturation and aggregation of excess adsorbent particles .Additionally, particle aggregation may occur at higher sorbent doses, which can reduce the effective surface area and cause overlap or blockage of the active sites. Furthermore, increased particle density can lead to diffusion restrictions and mass transfer resistance, reducing the accessibility of dye molecules to the deep internal pores. These results indicate that an optimal sorbent dose of 0.05 g provides the most efficient and economical dye removal, balancing high adsorption capacity with minimal material usage[33] .While removal efficiency (%) increased continuously with dosage, the adsorption capacity (qe, mg/g) decreased at higher dosages due to the reduced ratio of dye molecules to available active sites. Accordingly, the optimum dosage was defined as the point that balances high removal efficiency with relatively high qe, ensuring both effective pollutant removal and efficient adsorbent utilization, which is critical for economic feasibility.

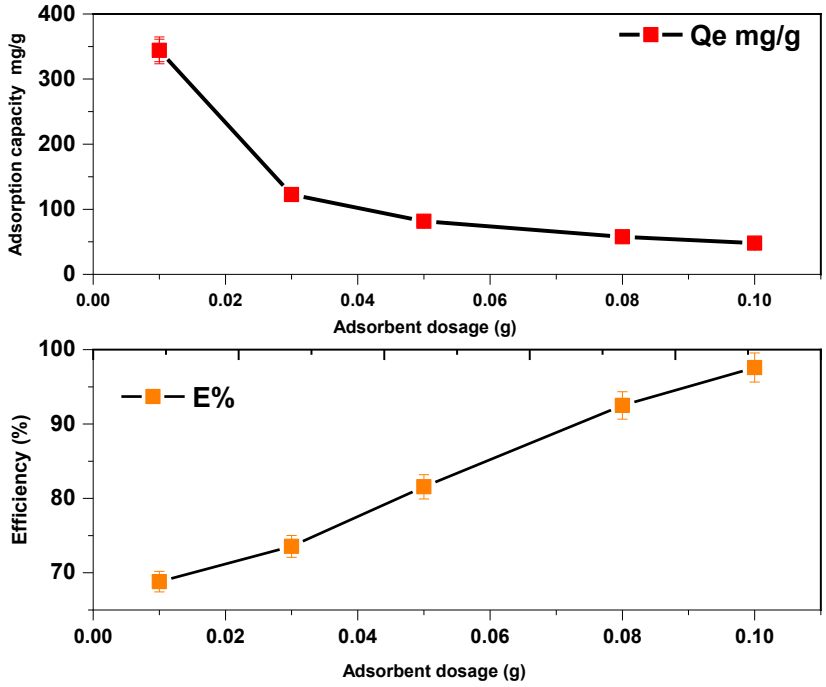

Figure 5. Effect of the weight of AC-HCl on the removal of MB dye

2.3. Comparison of the adsorption efficiency of natural, carbonized, and acid-activated castor leaves

This evaluation compared the adsorption capacities of three types of castor leaves: (1) raw natural leaves, (2) leaves thermally carbonized without chemical activation, and (3) acid-activated carbonized leaves Figure 6 . The untreated raw castor leaves showed limited adsorption capacity due to their low porosity and insufficient surface functionalization, which limited the number of active sites available for interaction with

dye molecules. In contrast, the carbonized leaves, produced through heat treatment at elevated temperatures, showed improved surface area and pore development. However, the adsorption capacity remained moderate because the lack of chemical activation resulted in fewer surface functional groups for dye binding[34]. The acid-activated carbonized leaves showed a significant improvement in adsorption efficiency. Hydrochloric acid treatment increased porosity and increased porosity and surface functionality, such as -OH, -COOH, and -C=O, to the adsorbent surface, as confirmed by FTIR analysis. These functional groups enhanced electrostatic interactions, hydrogen bonds, and π–π interactions with the dye molecules, resulting in superior dye removal efficiency. In summary, while natural and heat-treated castor leaves offer some adsorption potential, acid-activated carbonized leaves exhibit significantly better performance, making them a more effective and reliable biosorbent for dye removal from wastewater[31].

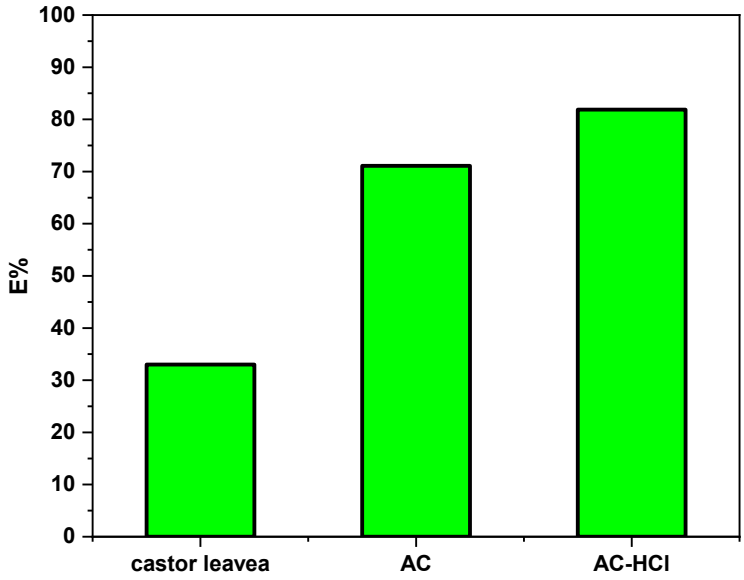

Figure 6. Comparison of the adsorption efficiency of natural, carbonized, and acid-activated carbon

2.4. Effect of initial concentration

Figure 7 illustrates the effect of initial methylene blue (MB) concentration (5–60 mg/L) on both the adsorption capacity (Qe, mg/g) and the removal efficiency (E%) of the hydrochloric acid–activated carbon derived from acid-activated Ricinus communis leaves. Adsorption Capacity Qe increased progressively from ~10 mg/g at 5 mg/L MB to ~94–95 mg/g at 60 mg/L MB.This continuous increase reflects the greater availability of dye molecules at higher concentrations, which enhances the driving force for mass transfer between the solution and the adsorbent surface. The nearly linear rise up to ~40 mg/L suggests efficient site utilization, while the tendency to approach a plateau beyond 50–60 mg/L indicates the progressive saturation of active sites.Removal Efficiency (E%) at low concentrations (5–10 mg/L) was exceptionally high (~98–99%), demonstrating that the available surface sites were more than sufficient to capture almost all dye molecules.As the concentration increased, E% gradually declined: ~95% at 20 mg/L, ~88% at 30 mg/L, ~83% at 40 mg/L, ~79% at 50 mg/L, and ~75% at 60 mg/L. This decline reflects the limited number of adsorption sites relative to the large number of dye molecules, leading to incomplete removal at higher concentrations. [35, 36].

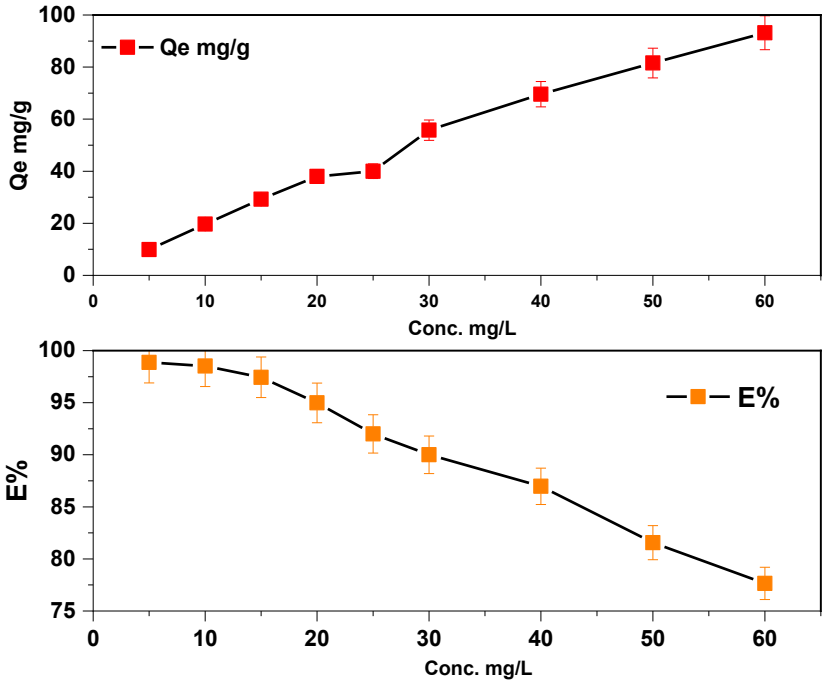

Figure 7. Effect of initial concentration by using AC-HCl

2.5. Regeneration and reuse of activated carbon adsorbents

Regeneration and reusing adsorbents are critical in enhancing cost-effectiveness and sustainability in wastewater treatment processes. This study conducted preliminary carbon adsorption experiments to evaluate the regeneration potential of an AC-HCl adsorbent prepared using different extracting agents. The adsorption capacity of methyl blue (MB^+) dye was assessed using three extracting agents: distilled water, 0.1 M hydrochloric acid (HCl), and 0.1 M sodium hydroxide (NaOH). Among the solutions tested, 0.1 M HCl showed the highest adsorption efficiency, demonstrating its superior ability to regenerate the adsorbent. As shown in Table 2. The adsorption-desorption cycle was repeated six times to evaluate its reuse potential further. As shown in Figure 8, the dye removal efficiency remained consistently high across the cycles, with only a slight decrease after the fifth regeneration. These results confirm the activated carbon robust regenerability and potential for repeated use without significant loss of adsorption performance, thus providing a practical and economically viable approach for dye removal in wastewater treatment applications [37]. Specifically, MB is a cationic dye, and the acidic environment created by HCl leads to protonation of negatively charged functional groups (e.g., $–COO^–$) on the adsorbent surface. This protonation decreases electrostatic attraction and may even impart a net positive charge to the adsorbent surface, resulting in electrostatic repulsion with MB^+ ions. Consequently, this facilitates efficient desorption and explains the observed effectiveness of HCl.

Table 2. Testing three eluents (H2O, HCl, NaOH) as a comparative the removal efficiency for the first regeneration cycle

Eluents	E%
HCl	81.66
NaOH	65
H2O	33%

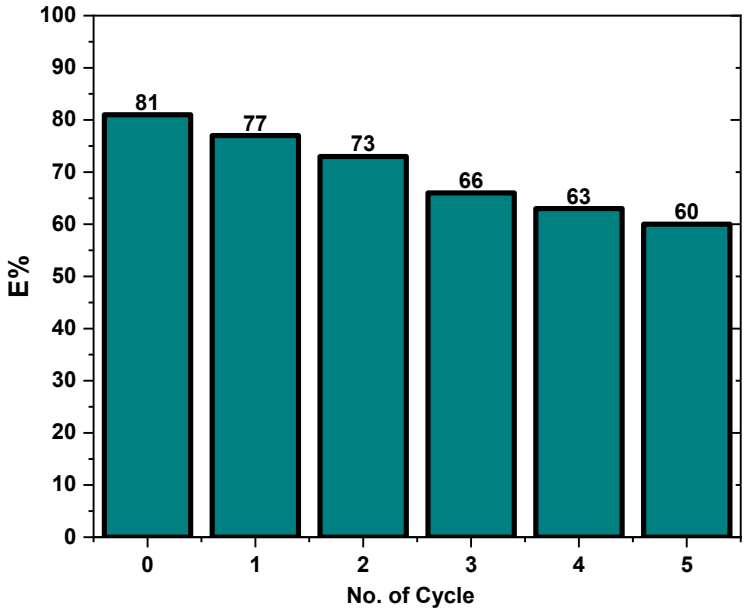

Figure 8. Regeneration and reuse of activated carbon adsorbents

3. Conclusion

This study highlights the promising potential of acid-activated Ricinus communis leaves as an efficient and low-cost biosorbent for removing dyes from aqueous solutions. Hydrochloric acid activation significantly improved the material's physical and chemical properties, including surface area, porosity, and the presence of functional groups that facilitate adsorption reactions. The effect of the sorbent dosage was systematically studied, revealing that increasing the dosage enhanced the dye removal efficiency up to the optimum (0.05 g). No significant increase in efficiency was observed beyond this dosage. This stability indicates saturation of the adsorption sites and the potential for competition between particles at high doses. Reuse tests demonstrated the material's stability and regeneration capacity. After multiple adsorption-desorption cycles using dilute hydrochloric acid as a regenerating agent, the sorbent maintained a high percentage of its initial removal efficiency, indicating its great potential for repeated use in practical applications. In summary, the acid-activated Ricinus communis leaves adsorbent demonstrated high adsorption capacity, chemical stability, and reusability, making it a sustainable and environmentally friendly option for treating dye-laden wastewater.

Conflict of interest

The authors declare no conflict of interest

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