



Fabrication of Activated Carbon-HKUST-1 MOF Nanocomposites for Enhanced Adsorption and Decontamination of Acidic and Basic Dyes in Wastewater

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ABSTRACT

The discharge of effluent containing synthetic dyes into aquatic systems presents significant environmental and health challenges, requiring modern treatment techniques. This study presents the production of an activated carbon–HKUST-1 (AC@HKUST-1) nanocomposite by an *in situ* ultrasonically assisted growth technique and its application for the simultaneous removal of cationic and anionic dyes. Structural characterisation (FTIR, XRD) demonstrated excellent crystallinity and retention of functional groups. Dynamic light scattering analysis revealed a Z-average hydrodynamic diameter of 26.11 nm and a polydispersity index of 1, indicating a relatively uniform size distribution. Batch adsorption experiments revealed fast absorption, with approximately 92% elimination of Rhodamine B (RhB) in 120 minutes and around 78% elimination of Methyl Orange (MO) in 180 minutes. The saturation sorption capacity attained was 276 mg g⁻¹ for RhB and 160 mg g⁻¹ for MO. Kinetic modelling demonstrated a superior fit to the pseudo-second order model (RhB: R² = 0.996; MO: R² = 0.992) in contrast to the pseudo-first order model (RhB: R² = 0.945; MO: R² = 0.932), thereby affirming chemisorption as the rate-limiting mechanism. Isotherm study revealed Langmuir monolayer adsorption for RhB (R² = 0.991; q_{max} ~300 mg g⁻¹) and Freundlich heterogeneity for MO (R² = 0.968; KF = 22, n = 1.9). The thermodynamic characteristics (ΔH° > 0; ΔG° -15 to -17 kJ mol⁻¹) indicate that the process is endothermic, spontaneous, and driven by entropy. These results underscore AC@HKUST-1 as a high-capacity, pH-adjustable, and sustainable adsorbent, demonstrating significant potential for scalable dye wastewater remediation.

Keywords: Dye adsorption; Metal-organic frameworks; Sustainability environment remediation; Wastewater treatment; Adsorption kinetics.

1. INTRODUCTION

The discharge of synthetic dyes into water bodies from industries such as textiles, leather, printing, and dye manufacturing represents a significant environmental concern. These effluents often contain organic colorants that are toxic, tumorigenic, and resistant to biodegradation (Al-Tohamy *et al.* 2022). Even at low concentrations, dyes hinder light penetration in aquatic ecosystems, disrupt photosynthesis, and produce undesirable colour, which can have detrimental effects on marine biota and pose potential risks to human health (Dutta *et al.* 2024). The global production of synthetic colorants exceeds seventy million metric tons annually, with the textile industry being a major contributor, responsible for consuming over ten thousand metric tons of these dyes (Chandanshive *et al.* 2020). Among the wide range of dyes, two primary types

cationic (basic) dyes, such as methylene blue and rhodamine B, and anionic dyes, including Congo red and methyl orange are particularly concerning due to their persistence and toxicity in aquatic environments (Holkar *et al.* 2016). These dyes, along with hazardous chemicals used during textile processing (e.g., sizing, softening, and whitening agents), are often released untreated into water bodies, leading to significant environmental pollution (Kishor *et al.* 2021; Parmar *et al.* 2022). Due to their distinct molecular structures and opposing charges, effective removal of both cationic and anionic dyes requires adsorbents capable of facilitating various non-covalent interactions, such as electrostatic attraction, π - π stacking, and hydrophobic interactions, across diverse pH conditions (Mnyango *et al.* 2025). Among the various methods for dye removal, adsorption is widely regarded

as a cost-effective and efficient approach. It offers high removal efficiencies even at low dye concentrations, making it an attractive option for wastewater treatment (Varshan *et al.* 2025). Activated carbon (AC), known for its high surface area and porous structure, has been extensively used in dye adsorption due to its relatively low cost. However, AC faces limitations such as pore blockage, slow adsorption kinetics for larger dye molecules, lack of selectivity, and instability under certain pH conditions (Pereira *et al.* 2003). To overcome these challenges, there has been a growing interest in composite adsorbents that combine the advantages of AC with other advanced materials (Katheresan *et al.* 2018).

Metal-organic frameworks (MOFs) are known for their high surface areas and tunable pore sizes, making them effective adsorbents for dye removal (Annamalai *et al.* 2022). Among MOFs, HKUST-1 (Cu-BTC) is particularly attractive for its high porosity and accessible Cu (II) sites, which are beneficial for dye adsorption applications (Zairov *et al.* 2025; Kabir *et al.* 2025). However, pure HKUST-1 faces limitations such as poor water stability, swelling, structural degradation, and high synthesis costs (Murugan *et al.* 2020). To address these issues, hybrid composites like AC-HKUST-1 have gained attention for their improved stability and enhanced adsorption performance (Loera *et al.* 2017).

To mitigate these limitations, combining AC with HKUST-1 in a nanocomposite structure offers a compelling alternative. In such composites, AC enhances the material's mechanical stability, increases porosity (especially meso- and macro-pores), and provides additional functional groups (e.g., -OH, -COOH) to improve dye interaction (Loera *et al.* 2017). HKUST-1 contributes to the composite's high affinity for dyes *via* its unsaturated metal sites and π - π interactions, creating a synergistic effect that improves adsorption kinetics, capacity, pH stability, and water resistance. Recent studies have demonstrated successful application of AC-MOF composites for dye removal, showing higher removal efficiencies compared to either material alone (Zeng *et al.* 2018; Jimenez *et al.* 2025). Moreover, modifications such as incorporating magnetic HKUST-1 or combining MOFs with metal oxides have further enhanced dye removal capacity, particularly for basic dyes like methylene blue (Qiao *et al.* 2023). This study aims to explore the potential of an activated carbon-HKUST-1 composite, synthesized *via* an *in situ* ultrasonically assisted growth method, for the enhanced adsorption of both cationic and anionic dyes. By leveraging the combined benefits of AC and HKUST-1, the composite aims to offer a more efficient and versatile solution to dye wastewater remediation, addressing the limitations observed in previous AC-HKUST-1 studies.

2. MATERIALS AND METHODOLOGY

2.1 Materials and Reagents

Coconut shells were procured from a local market and used as the precursor for activated carbon (AC). Potassium hydroxide (Merck), copper (II) nitrate trihydrate, and 1,3,5-benzenetricarboxylic acid (Sigma-Aldrich) were employed as chemical reagents. Ethanol, hydrochloric acid (HCl), and the dyes Methyl Orange and Rhodamine B (Fig. 1) were obtained from Sisco Research Laboratories Pvt. Ltd. (SRL), India. Throughout the experimental work, distilled water was employed to ensure the reliability and reproducibility of results.

2.2 Synthesis of Activated Carbon

The collected coconut shells were initially copiously rinsed with tap water and subsequently with distilled water, to ensure the removal of surface-bound impurities. They were then oven-dried at 80 °C for 24 hours. The dried material was crushed and sieved to obtain particles smaller than 2 mm. Carbonization was performed in a muffle furnace at 500 °C for 2 hours (Sujiono *et al.* 2022).

The prepared char was incorporated with KOH at a weight ratio of 1:3, mixed with a small quantity of distilled water to form a paste, and dried at 105 °C. The dried mixture was then treated to induce activation at 500 °C under a nitrogen atmosphere for one hour. After activation, the material was washed with 0.5 M HCl and repeatedly rinsed with distilled water until neutrality was reached (Yuliusman *et al.* 2020). The activated carbon obtained was dried in an oven at 100 °C and subsequently stored in airtight containers.

2.3 Synthesis of HKUST-1 MOF

To form Solution A, copper (II) nitrate trihydrate (2 mmol) was solubilized by adding 50 mL of distilled water. Separately, BTC (1 mmol) was prepared in a 1:1 (v/v) ethanol-water mixture containing 5 vol% acetic acid, yielding Solution B. This was gradually added to Solution A under continuous stirring, and the mixture was held quiescent at ambient temperature for 24 hours (Sun *et al.* 2019). The resulting blue precipitate was isolated by filtration, thoroughly rinsed sequentially with ethanol and water, and subsequently dried at 80 °C overnight.

2.4 Fabrication of Activated Carbon- HKUST-1 Composite

In situ Ultrasonically Assisted Growth Method was used to fabricate the composite. Activated carbon (0.5 g) was dispersed in 50 mL ethanol: water (1:1 v/v) by ultrasonication for 30 minutes. Copper nitrate (2 mmol) was added to the suspension and stirred for 30 minutes to facilitate ion adsorption on the AC surface.

Under stirring BTC (1 mmol) in 50 mL ethanol: water was added, and the solution was ultrasonicated for another 30 minutes (40 kHz/60W). The entire mixture was transferred into a Teflon-lined autoclave and heated at 100 °C for 8 hours (Zeng *et al.* 2018; Jimenez-Lopez *et al.* 2025; Li *et al.* 2008). Once the solution reached room temperature, the AC@HKUST-1 composite was collected *via* centrifugation, washed multiple times with ethanol followed by water, and dried at 100 °C.

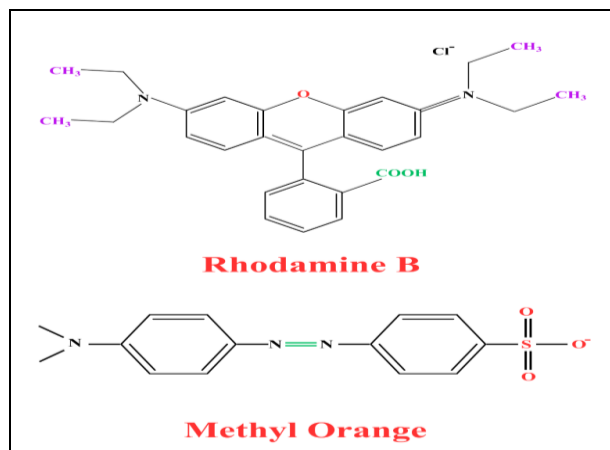


Fig. 1: Molecular structure of dyes

2.5 Characterization of Activated Carbon-HKUST-1 MOF (AC@HKUST-1) Nanocomposite

The prepared nanocomposite (AC@HKUST-1) was characterized using X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), dynamic light scattering (DLS), and scanning electron microscopy (SEM) with energy dispersive X-ray spectroscopy (EDX). Crystallite size and phase purity were analyzed using XRD, functional group interactions were confirmed through FTIR, and DLS provided the particle size distribution and polydispersity index (PDI) of AC@HKUST-1. Scanning electron microscopy coupled with EDX was employed to observe the surface morphology and elemental composition of the materials at a microscopic level.

2.6 Adsorption of Acidic and Basic Dyes

The synthesized AC@HKUST-1 nanocomposite was evaluated for Methyl Orange (MO) and Rhodamine B (RhB) sequestration *via* batch adsorption experiments. In each experiment, 25 mg of the adsorbent was introduced into 100 mL of dye solution (10 mg L⁻¹) in a 250 mL conical flask, and the pH was adjusted to the specified level with 0.1 M HCl/NaOH. The resulting suspension was set on an orbital shaking incubator maintained at 37 °C and stirred at 200 RPM for 3 hours to facilitate homogeneous mixing and interaction between dye molecules and adsorption sites. Aliquots were withdrawn at 30-minute intervals, the mixture was

clarified by filtration through Whatman filter paper of 0.45 µm pore size, and subsequently characterized using a UV-Vis spectrophotometer. This procedure enabled the evaluation of adsorption kinetics and efficiency under varying experimental conditions.

2.7 Kinetic Studies of Dye Adsorption

Kinetic adsorption experiments were conducted using 50 mg of AC@HKUST-1 MOF adsorbent in 100 mL of methyl orange and rhodamine B solutions, each at a concentration of 50 mg/L, at a temperature of 25 ± 1 °C. The solid-liquid ratio was maintained at 0.05 g/L. The mixture samples were subjected to agitation at a uniform speed of 150 rpm for predetermined intervals of 5, 15, 30, 60, 120, and 180 min to probe the time-dependence of adsorption efficiency. After each interval, suspensions were centrifuged (5000 rpm, 10 min) and filtered (0.45 µm). UV-Vis spectrophotometry determined residual dye concentrations at λ_{max} 464 nm for MO and 554–556 nm for RhB (Chennouf *et al.* 2017). To ensure accuracy and reproducibility, each experiment was executed in triplicate, with the averaged observations subsequently applied to kinetic modeling and analysis of the adsorption process.

Adsorption isotherms were studied by varying the initial concentrations of MO and RhB (10–200 mg/L) at 25, 35, and 45 °C. Each 250 mL flask contained 50 mg of AC@HKUST-1 MOF in 100 mL of dye solution, agitated at 150 rpm until equilibrium (Wu *et al.* 2017; Dada *et al.* 2019; Wijaya *et al.* 2021).

To evaluate the role of solution pH in dye adsorption, experiments were performed at pH values of 3, 5, 7, 9, and 11. pH of the dye solutions was pre-adjusted with 0.1 M HCl or 0.1 M NaOH before adding the adsorbent (AC@HKUST-1 MOF). The experiments were conducted with an adsorbent dosage of 50 mg in 100 mL of dye solution (50 mgL⁻¹) at 25 °C and an initial dye concentration of 50 mgL⁻¹. This procedure was adapted from (Sabouni *et al.* 2019). Control sample, without adsorbent or dye was maintained using distilled water to account for and eliminate non-adsorptive losses. All experiments were executed in triplicate, and accuracy reported is the mean across replicates.

2.8 Adsorption Calculations

The adsorption capacity at time *t* (*q_t*) and equilibrium (*q_e*) was quantified from dye concentrations as:

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

Removal efficiency (%) was computed by:

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

where C_0 and C_e represent initial and equilibrium dye concentrations and at a time (t) (mg/L), V is volume (L) of the solution, and m is mass of AC@HKUST-1 MOF (g).

2.9 Kinetic Models

Kinetic data were analyzed by fitting them to the pseudo-first order (PFO) model (Lagergren, 1898) and the pseudo-second order (PSO) model to describe adsorption rates.

$$\text{PFO: } \log(q_e - q_t) = \log(q_e) - \frac{K_1}{2.303} t \quad \dots (3)$$

$$\text{PSO: } \frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad \dots (4)$$

where K_1 (min^{-1}) and K_2 ($\text{g mg}^{-1} \text{min}^{-1}$) are kinetic constants. The best-fitting model was identified based on correlation coefficients (R^2) and error analysis (RMSE, χ^2), 2.303 is the logarithmic conversion factor.

2.10 Isotherm Models

Equilibrium adsorption data were fitted to Langmuir and Freundlich isotherms, representing monolayer adsorption capacity and surface heterogeneity, respectively.

$$\text{Langmuir: } \frac{C_e}{q_e} = \frac{1}{q_{\max} K_L} + \frac{C_e}{q_{\max}} \quad \dots (5)$$

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

where $q_{\max}$ (mg g^{-1}) denotes the monolayer adsorption capacity, K_L (L mg^{-1}) is the Langmuir constant, and the Freundlich coefficients K_F (mg g^{-1}) and $1/n$ characterize surface heterogeneity.

2.11 Thermodynamic Parameters

The distribution coefficient (K_d) was calculated as $K_d = q_e/C_e$. Thermodynamic parameters were derived using:

$$\Delta G^0 = -RT \ln K_d \quad \dots (7)$$

$$\ln K_d = \frac{\Delta H^0}{RT} + \frac{\Delta S^0}{R} \quad \dots (8)$$

The absolute temperature (T K) and universal gas constant ($R = 8.314 \text{ J mol}^{-1} \text{K}^{-1}$) were applied in thermodynamic calculations. The Gibbs free energy change (ΔG^0) assessed adsorption spontaneity, while enthalpy (ΔH^0) and entropy (ΔS^0) were obtained from temperature-dependent studies to elucidate the adsorption mechanism. The reliability of model fitting

was assessed using correlation coefficients and error analyses to confirm methodological accuracy.

3. RESULTS AND DISCUSSION

3.1 Fourier Transform Infrared Spectrum of the AC@HKUST-1 MOF

The FTIR spectrum of the AC@HKUST-1 MOF composite revealed key vibrational features that confirm the successful hybridization of activated carbon with the copper-based HKUST-1 framework (Fig. 2). A broad absorption peak centered at $3335\text{--}3307 \text{ cm}^{-1}$ is ascribed to O–H stretching vibrations, which can be attributed to surface hydroxyl groups and adsorbed water molecules commonly associated with carbonaceous and MOF materials (Huang *et al.* 2018; Jahan and Feni, 2022). Distinct peaks at 2924 and 2848 cm^{-1} correspond to asymmetric and symmetric stretching vibrations of aliphatic C–H bonds, reflecting residual organic moieties in the activated carbon matrix (Tsai *et al.* 2004; Shin 2021). A weak band observed at 2378 cm^{-1} is attributed to the stretching vibration of O=C=O, which is characteristic of carbon dioxide from the ambient atmosphere (Barakat *et al.* 2020). The absorption peak at 2102 cm^{-1} corresponds to the C≡C stretching vibration, indicating the presence of an alkyne group (Prasad *et al.* 2011). The peak at 1908 cm^{-1} arises from conjugated carbonyl vibrations, such as in anhydrides or α , β -unsaturated ketones, along with possible aromatic overtones. Additionally, the peak at 1622 cm^{-1} corresponds to C=O stretching of carboxylic acids, evidencing oxygenated groups on the activated carbon surface (Pallarés *et al.* 2018; Freitas *et al.* 2019; Mojoudi *et al.* 2019). The peaks observed at 1647 , 1622 , and 1437 cm^{-1} correspond to the symmetric and asymmetric stretching modes of the C=O bond. In the spectra of AC and HKUST-1 MOF, a peak around 1102 cm^{-1} is attributed to C–O stretching vibrations, which is also present in the AC-HKUST-1 spectrum (Azad *et al.* 2016). These features indicate the existence of oxygen-rich functional groups on the activated carbon, which provide anchoring sites for MOF crystallization. In the fingerprint region, the peak at 1026 cm^{-1} corresponds to C–O stretching vibrations of alcohols or ethers, while the shoulder at 950 cm^{-1} is consistent with C–H bending modes (Smith, 2022). The stretching peaks corresponding to the Cu–O bond were observed at 732 cm^{-1} , with the FTIR peak at this wavelength confirming the formation of a metal-linker bond between Cu and BTC (Klęba *et al.* 2025). The sharp absorption at 707 cm^{-1} is attributed to the out-of-plane bending vibration of aromatic C–H bonds (Hadjiivanov *et al.* 2020). The coexistence of MOF-related (Cu–O, aromatic) and carbon-related (–OH, –COOH, C–O) vibrations highlights the synergistic integration, which is expected to enhance adsorption, stability, and catalytic performance of the composite in environmental applications.

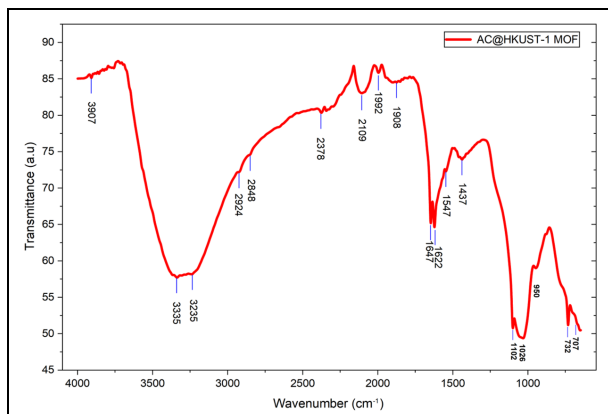


Fig. 2: FTIR spectrum of activated carbon-HKUST-1 MOF nanocomposite

3.2 XRD Patterns of Activated Carbon@HKUST-1 MOF

The crystalline structure of the Activated Carbon@HKUST-1 MOF composite was analyzed using XRD with triple-axis ω - 2θ scans conducted on a PANalytical XRD system. The XRD spectrum of synthesized Activated Carbon@HKUST-1 MOF is shown in Fig. 3. The peaks at 5° , 6.7° , 9.5° , 11.6° , 13.4° and 17.4° correspond to the (111), (200), (220), (222), (400) and (511) crystal planes of HKUST-1, confirming its well-defined crystalline structure and high symmetry (Rozaini *et al.* 2023; Klęba *et al.* 2025; Zeng *et al.* 2018). While other peaks at 26.1° confirm the presence of activated carbon, corresponding to the (002) crystallographic plane (Kalagatur *et al.* 2017). The broad background hump in the range of 20 – 30° is ascribed to the amorphous nature of the activated carbon matrix, which coexists with the ordered MOF lattice (Ghafuri *et al.* 2020). These results confirm the successful incorporation of HKUST-1 onto activated carbon, where the crystalline peaks of the MOF are preserved, and the carbon contributes to enhanced stability and textural properties suitable for adsorption and catalytic applications.

3.3 Dynamic Light Scattering (DLS) of AC@HKUST-1 MOF

Dynamic Light Scattering was employed to assess the hydrodynamic size and dispersion characteristics of the AC@HKUST-1 MOF particles in aqueous suspension. The measured Z-average size was approximately 26.11 nm, indicating that the particles are well within the nanoscale range, as shown in Fig. 4. The PDI was calculated to be 1, suggesting a relatively uniform size distribution within the sample. This PDI value supports the presence of a homogeneous nanoparticle population, which is crucial for ensuring reproducible performance in nanoscale applications. The mean count rate was recorded at 1546 kcps, indicating a

strong scattering signal and high particle concentration within the measured range. The baseline index was determined to be 0.0, further confirming the presence of a monodisperse nanoparticle suspension.

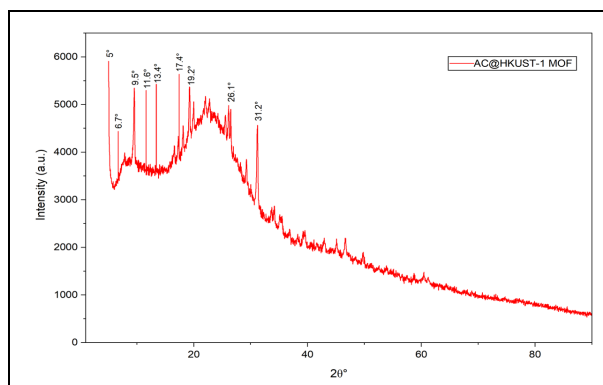


Fig. 3: XRD pattern of activated carbon@HKUST-1 MOF

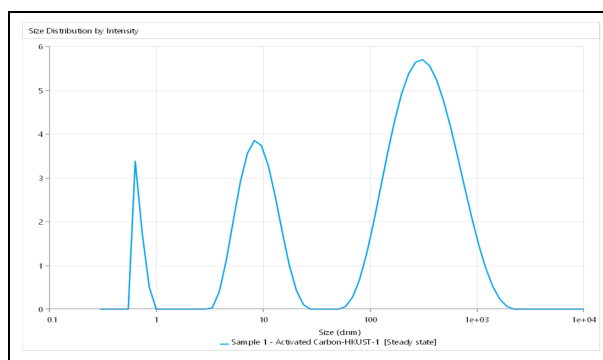


Fig. 4: DLS analysis graph of synthesized activated carbon@HKUST-1 MOF

3.4 Dye Adsorption Performance of AC@HKUST-1 MOF

Wastewater from chemical and textile industries contains toxic dyes. Among the different treatment techniques, adsorption is widely recognized as widely used due to its straightforward operation, cost-effectiveness, and effective dye pollutant removal (Rajak *et al.* 2017). Batch adsorption studies were conducted to evaluate the performance of the synthesized AC@HKUST-1 nanocomposite in removing two structurally distinct dyes, MO and RhB from aqueous solutions. A 10 mg/L dye solution (100 mL) was mixed with 25 mg of the adsorbent, and the adsorption process was carried out at an agitation speed of 200 rpm and a constant temperature of 37°C . Aliquots were withdrawn at 30-minute intervals, and the residual dye concentration was quantified using UV-Vis spectrophotometry. The calibration curves for both dyes were constructed to quantify dye concentrations over time (Fig. 5a, 5b). The R^2 values were 0.9879 for RhB and 0.9736 for MO, indicating excellent linearity and a reliable correlation between absorbance and dye concentration. These high

R^2 values demonstrate the precision and effectiveness of the spectrophotometric method for dye quantification.

The FTIR spectra of AC@HKUST-1 before and after adsorption of RhB and MO provided insights into the surface functional groups involved in the adsorption process (Fig. 5c). The spectrum of AC@HKUST-1 before adsorption, showed peaks at 3243 cm^{-1} , 2848 cm^{-1} , and 1437 cm^{-1} , corresponding to hydroxyl, C–H stretching, and C=O bending vibrations, respectively, confirming the presence of surface oxygen functionalities and organic groups (Azad *et al.* 2016; Tsai *et al.* 2004; Shin 2021). The peak at 1024 cm^{-1} was attributed to C–O stretching vibrations (Smith, 2022). After the adsorption of Rhodamine B significant shifts were observed in the peaks at 3260 cm^{-1} and 2915 cm^{-1} , indicating interactions between the dye molecules and the surface hydroxyl and carbonyl groups (Isik, 2024). New peaks at 1440 cm^{-1} and 1000 cm^{-1} appeared, suggesting the formation of electrostatic interactions and hydrogen bonding between the dye and the adsorbent surface. Similarly, after MO adsorption, the peak at 2907 cm^{-1} , attributed to C–H stretching, became less intense, reflecting the interaction between the dye and the surface. The peaks at 1647 cm^{-1} and 1152 cm^{-1} also showed notable changes, confirming the adsorption of MO onto AC@HKUST-1 (Sayed *et al.*, 2024). These FTIR changes indicate that both RhB and MO interact with the surface functional groups of AC@HKUST-1, forming hydrogen bonds and electrostatic interactions that play a crucial role in the adsorption process. The findings confirm that AC@HKUST-1 is an effective adsorbent for the removal of both anionic and cationic dyes from aqueous solutions.

To further understand the adsorption behavior, the pH of the Point of Zero Charge (pH_{PZC}) for AC@HKUST-1 was determined. The pH_{PZC} of AC@HKUST-1 was found to be approximately 6.48, (Fig. 5d) which marks the transition from positive to negative surface charge. This finding is significant for dye adsorption applications, where the surface charge of the adsorbent dictates its ability to attract and adsorb various dyes. At $\text{pH} < \text{pH}_{\text{PZC}}$, the positively charged surface of AC@HKUST-1 favors the adsorption of anionic dyes (like Methyl Orange). At $\text{pH} > \text{pH}_{\text{PZC}}$, the surface is negatively charged, making it favorable for the adsorption of cationic dyes (like Rhodamine B). This pH_{PZC} determination offers insight into optimizing pH conditions for effective dye removal processes.

3.5 SEM coupled with EDX of AC@HKUST-1 MOF

The SEM-EDX analysis of AC@HKUST-1 MOF provides valuable insights into both the structural and elemental characteristics of the material. The SEM images (6a and 6b) reveal the morphology of the AC@HKUST-1 composite, showing nanosized crystalline aggregates with rough and irregular surfaces.

At magnifications of 8.23 KX and 3.44 KX, the images highlight the porous structure and granular arrangement of the particles. The irregularity in shape and surface roughness suggests that the composite material possesses a high surface area, which is desirable for applications such as catalysis and adsorption. The SEM images further indicate that the particles are well-dispersed, providing a large contact area for interactions with reactants or adsorbates. The EDX spectrum (6c) reveals the elemental composition of the composite, identifying key peaks for copper (Cu), oxygen (O), and carbon (C), which are essential components of the AC@HKUST-1 structure. The sharp peaks for copper and oxygen validate the successful incorporation of the HKUST-1 framework, based on copper, onto the activated carbon substrate. The carbon peaks further support the presence of activated carbon. This combined SEM-EDX analysis offers essential structural and compositional details, reinforcing the potential of AC@HKUST-1 as a versatile material for various functional applications.

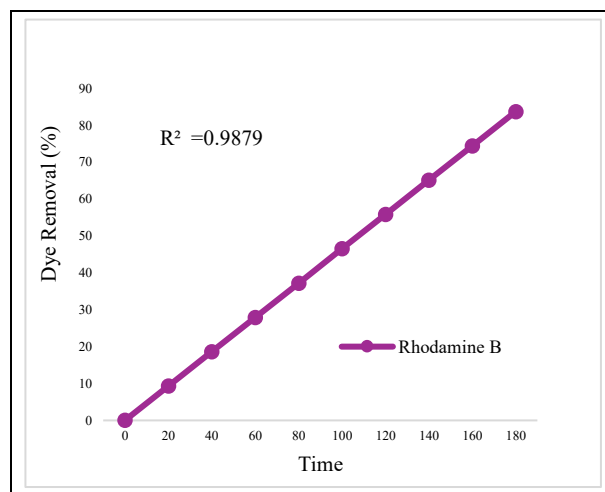


Fig. (5a): Calibration curve for rhodamine B dye removal

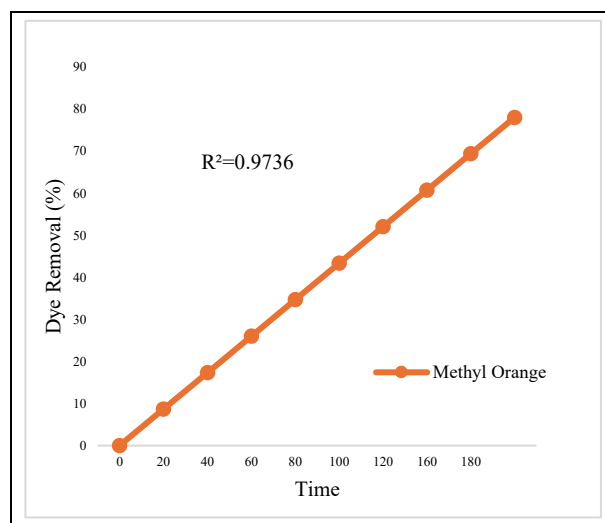


Fig. (5b): Calibration curve for methylene orange dye removal

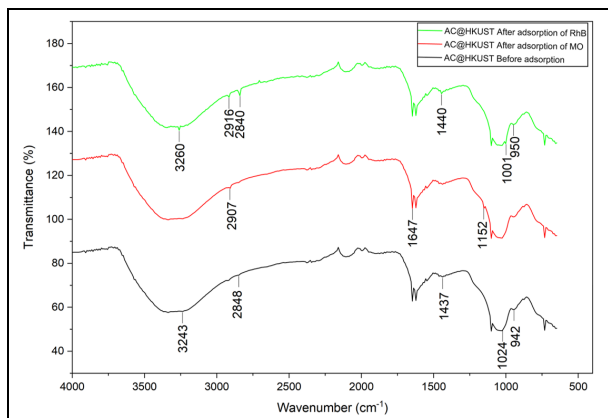


Fig. (5c): FTIR-ATR spectra of AC@HKUST-1 before and after RhB and MO adsorption

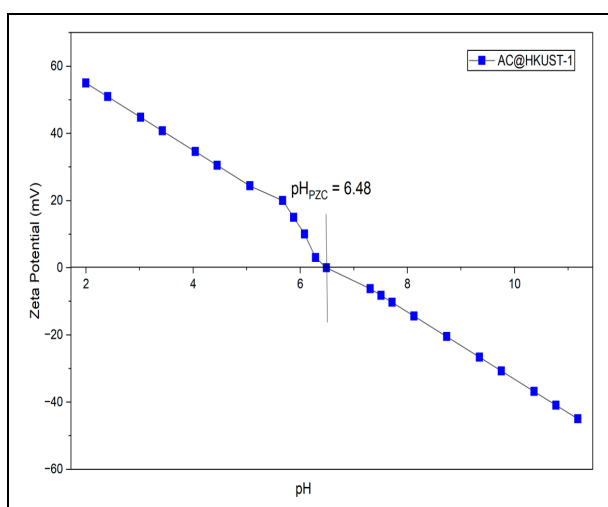


Fig. (5d): Effect of pH on the zeta potential of AC@HKUST-1 and determination of pH_{PZC}

Table 1. Kinetics of RhB and MO adsorption on AC–HKUST-1 ($C_0 = 50 \text{ mg L}^{-1}$; dose = 0.05 g; $V = 100 \text{ mL}$; 25°C)

Time (min)	C_0 (RhB, mg L^{-1})	q_e (RhB, mg g^{-1})	Removal (%)	C_0 (MO, mg L^{-1})	q_e (MO, mg g^{-1})	Removal (%)
5	35	30.0	30	43	14.0	14
15	25	50.0	50	36	28.0	28
30	18	64.0	64	30	40.0	40
60	12	76.0	76	23	54.0	54
120	4	92.0	92	14	72.0	72
180	4	92.0	92	11	78.0	78

3.6 UV–Vis Absorption Spectra of Dyes

The absorbance spectra for RhB (Fig. 7a) exhibited a prominent band at 565 nm, characteristic of its chromophore. Over time, the intensity of this band decreased significantly, indicating the progressive removal of the dye. By 180 minutes, the absorbance at

λ_{max} had decreased by over 60%, confirming substantial dye removal. Similarly, the UV-Vis spectra of MO (Fig. 7b) showed bands at 220 nm and 465 nm, indicative of its complex aromatic structure. Both bands exhibited a steady decrease in intensity, and the colour of the solution changed from orange to pale yellow, confirming significant dye removal.

3.7 Adsorption Kinetics

Adsorption kinetics for the AC–HKUST-1 nanocomposite was studied using MO and RhB at room temperature with an initial dye level of 50 mg L^{-1} . The influence of contact time on uptake capacity and total removal efficiency was assessed. The kinetic experiments showed a rapid initial uptake within 30 minutes for both dyes (Table 1; Fig. 8a, b, c). Both dyes exhibited a sharp increase in adsorption capacity throughout the first 30 minutes, indicating a rapid surface uptake and strong affinity of the adsorbent. Thereafter, the adsorption rate progressively declined, reflecting a transition from surface interaction to gradual intraparticle diffusion and site saturation. Equilibrium was achieved more quickly for RhB, with $\sim 92\%$ removal observed within 120 minutes, whereas MO adsorption proceeded slowly, attaining $\sim 78\%$ removal only after 180 minutes (Fig. 8d).

Rapid adsorption kinetics of RhB can be attributed to stronger electrostatic interactions between its cationic molecules and the negatively charged functional sites of AC–HKUST-1, which enhance the adsorption affinity and facilitate faster diffusion compared to MO. To contextualize these findings, the kinetic behavior was compared with previously reported nanostructured and MOF-based adsorbents (Table 1). These results highlight the potential of AC–HKUST-1 MOF as an effective multifunctional adsorbent for cationic and anionic dyes, combining rapid kinetics with high removal efficiency. The distinct kinetic profiles of RhB and MO further emphasize the importance of dye–adsorbent interactions, solution chemistry, and molecular structure in dictating adsorption performance.

3.8 Adsorption Isotherms (25°C)

Equilibrium adsorption of RhB and MO onto AC–HKUST-1 was examined over a baseline concentration range of $10\text{--}200 \text{ mg L}^{-1}$ (Table 2; Fig. 9a, b, c). The adsorption capacity (q_e) increased with increasing C_0 , approaching saturation at higher concentrations, reflecting progressive occupation of active sites. At $C_0 = 200 \text{ mg L}^{-1}$, maximum uptake reached 276 mg g^{-1} for RhB and 160 mg g^{-1} for MO, indicating a stronger affinity of the composite toward RhB. This difference is attributed to $\pi\text{--}\pi$ stacking between RhB aromatic rings and graphitic domains of the AC support, together with electrostatic interactions

between cationic RhB and oxygenated functional groups. In contrast, MO adsorption was lower, consistent with the greater pH sensitivity of its anionic species.

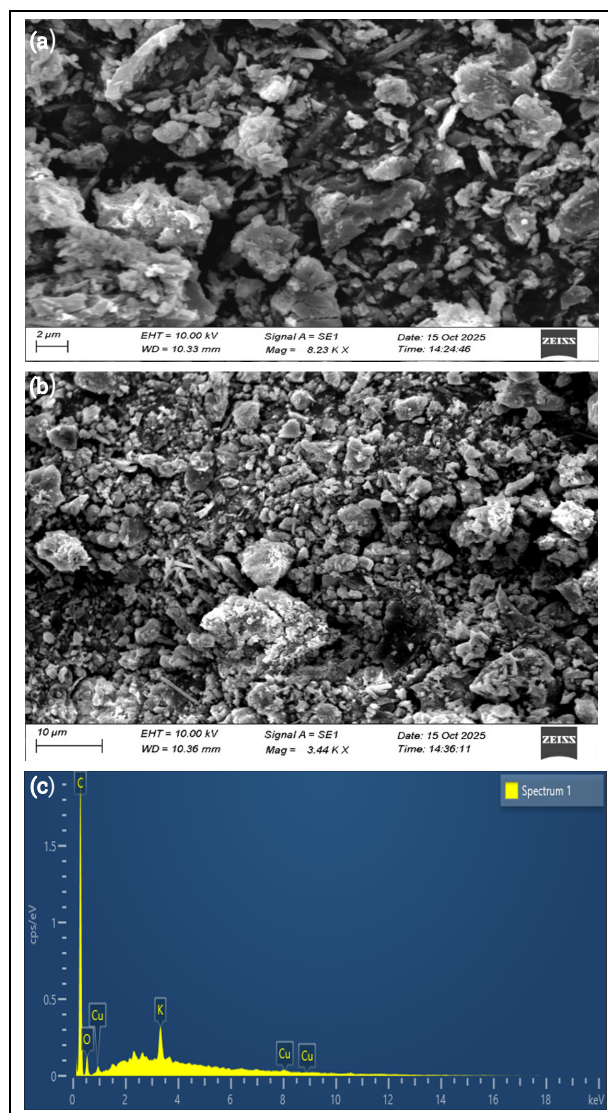


Fig. 6: (a) SEM image of AC@HKUST-1 MOF at 8.23 KX magnification (b) SEM image of AC@HKUST-1 MOF at 3.44 KX magnification and (c) EDX spectrum of AC@HKUST-1

Table 2. Isotherm data (25 °C dose = 50 mg, V = 100 mL)

C_0 (mg L ⁻¹)	C_e (RhB)	q_e (RhB, mg g ⁻¹)	C_e (MO)	q_e (MO, mg g ⁻¹)
10	0.8	18.4	2.2	15.6
20	2.5	35.0	5.5	29.0
40	7.5	65.0	13.0	54.0
80	18.0	124.0	32.0	96.0
120	32.0	176.0	57.0	126.0
160	50.0	220.0	88.0	144.0
200	62.0	276.0	120	160.0

Table 3. Isotherm model parameters (25 °C; linear forms)

Dye	Langmuir q_{max} (mg g ⁻¹)	K_L (L mg ⁻¹)	R^2 (Langmuir)	Freundlich KF	n	R^2 (Freundlich)
RhB	~300	0.12	0.991	42	2.4	0.965
MO	190	0.05	0.952	22	1.9	0.968

Table 4. Effect of pH on removal of dyes (dose-50 mg; V-100 mL; 25 °C)

pH	Removal (%) RhB	Removal (%) MO
3	58	78
5	72	84
7	88	79
9	92	65
11	95	57

Table 5. Dosage effect on removal (V = 100 mL; 25 °C)

Dose (mg)	Removal (%) RhB	Removal (%) MO
10	45	36
50	90	80
100	96	90
150	98	94
500	99	96

Table 6. Temperature effect and thermodynamic parameters (dose = 50 mg; V = 100 mL)

T (°C)	q_e (RhB, mg g ⁻¹)	q_e (MO, mg g ⁻¹)	ΔG° (RhB, kJ mol ⁻¹)	ΔG° (MO, kJ mol ⁻¹)
25	90	74	-15.1	-13.4
35	100	82	-16.2	-14.3
45	110	88	-17.3	-15.0

Isotherm modeling (Table 3) revealed distinct adsorption behaviours. For RhB, the Langmuir model offered the best fit ($R^2 = 0.991$), with a calculated monolayer capacity of ~ 300 mg g⁻¹ and $K_L = 0.12$ L mg⁻¹, suggesting uniform adsorption on homogeneous sites of the composite. Conversely, MO was aligned more strongly with the heterogeneous-surface assumption of the Freundlich model ($R^2 = 0.968$, KF = 22, n = 1.9), indicating adsorption on heterogeneous surfaces with variable binding affinities. The Freundlich parameter n = 1.9 suggests favorable adsorption. This finding is consistent with the notion of a non-uniform distribution of adsorption energies for anionic dyes, where multiple adsorption sites with varying affinity contribute to the overall adsorption process (Anuar *et al.* 2019). The present composite exceeds these benchmarks, underscoring the synergistic role of activated carbon in enhancing adsorption. Overall, these results confirm AC–HKUST-1 as a versatile adsorbent with distinct adsorption mechanisms for cationic and anionic dyes, guided by molecular structure and surface chemistry.

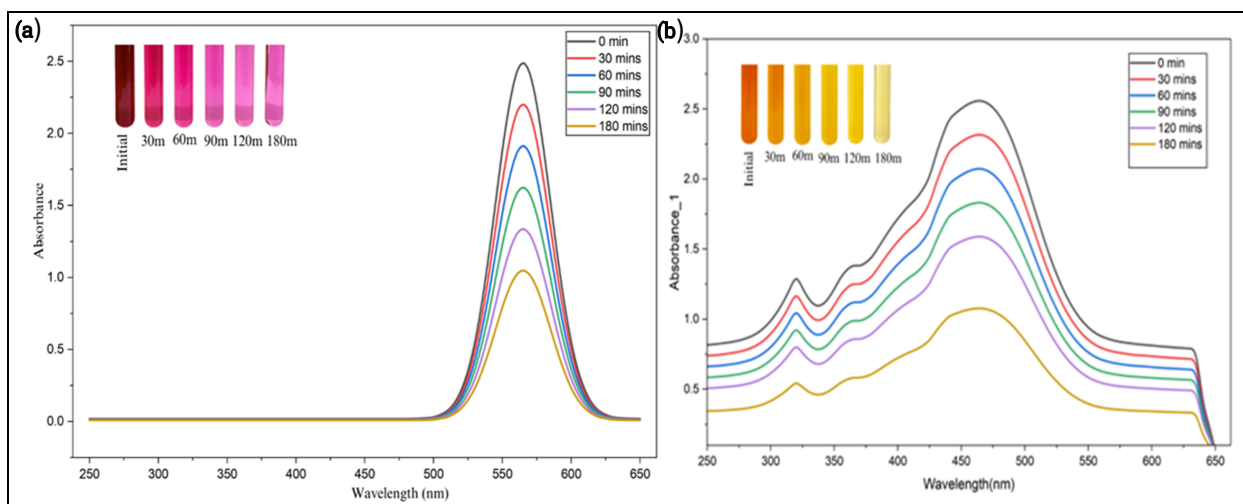


Fig. 7: UV-vis absorption spectra of (a) RhB dye and (b) MO dye during treatment with MOF nanocomposite over 0–180 min

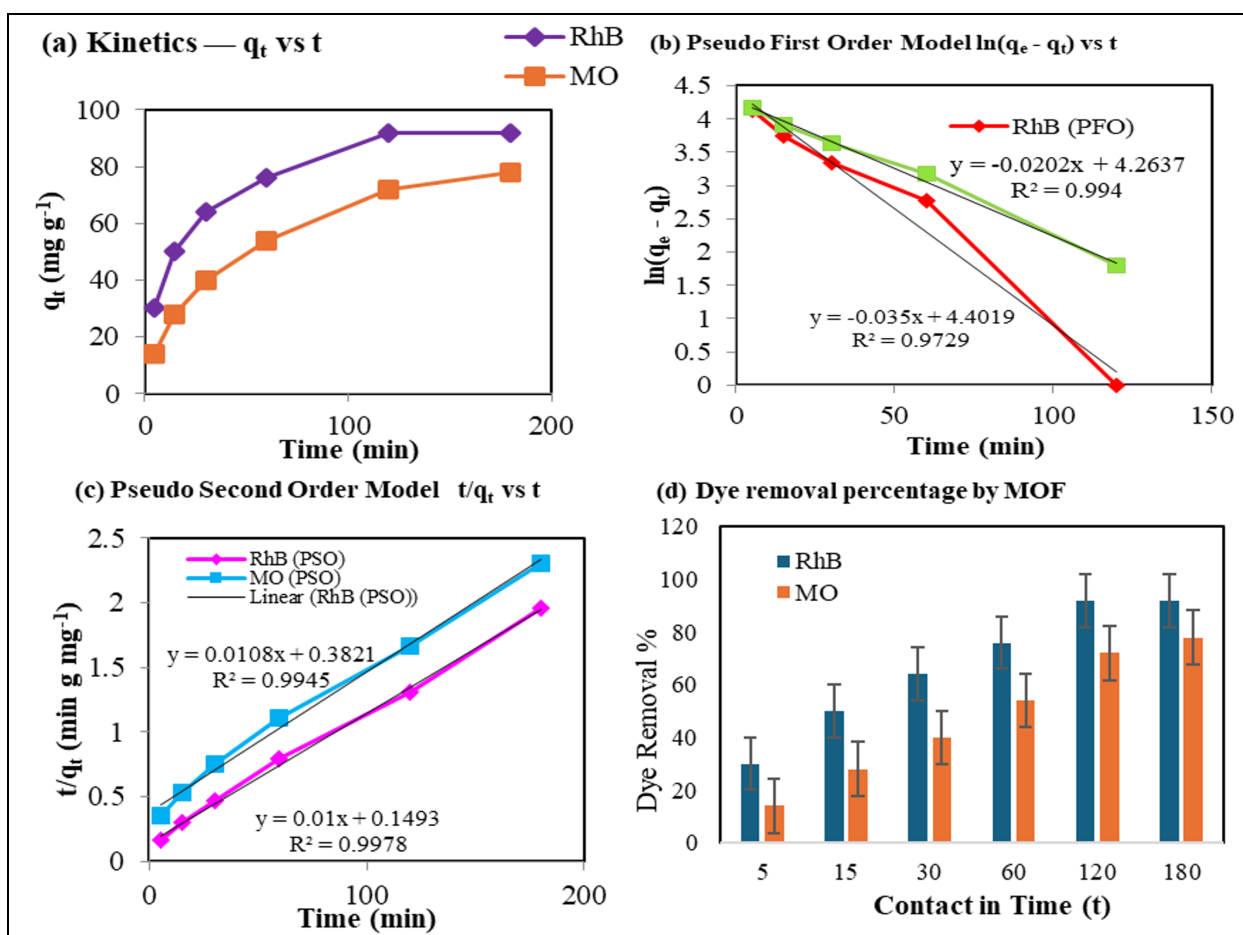


Fig. 8: Adsorption kinetics and kinetic modeling of RhB and MO dyes on the synthesised adsorbent (a) Adsorption kinetics (q_t vs. Time) (b) Pseudo-first-order kinetic model (c) Pseudo-second-order kinetic model and (d) Dye removal percentage

The pH of the solution plays a crucial role in the adsorption of dyes on AC-HKUST-1, influencing both the surface charge of the adsorbent and the ionization of the dye molecules. In addition, various interactions, including π - π interactions, n - π interactions, hydrogen bonding, and hydrophobic interactions, contribute to the

overall adsorption process in aqueous solutions (Li *et al.* 2013). The removal efficiency of MO at different pH levels (ranging from pH 3 to pH 11) is presented in Table 4, Fig. 10a. The maximum MO removal (~84%) occurred at pH 5, which is slightly acidic. However, as the pH increased to pH 11, the removal efficiency

decreased significantly to approximately 57%. This decline can be attributed to electrostatic repulsion between the negatively charged surface of AC-HKUST-1 at higher pH and the anionic nature of the MO dye. At pH 5, the surface of the adsorbent is likely positively charged, which enhances the attraction of the negatively charged MO molecules. At higher pH, the surface becomes negatively charged, leading to repulsion with the anionic dye, thereby reducing adsorption efficiency (Serban *et al.* 2023; Shakly *et al.* 2022).

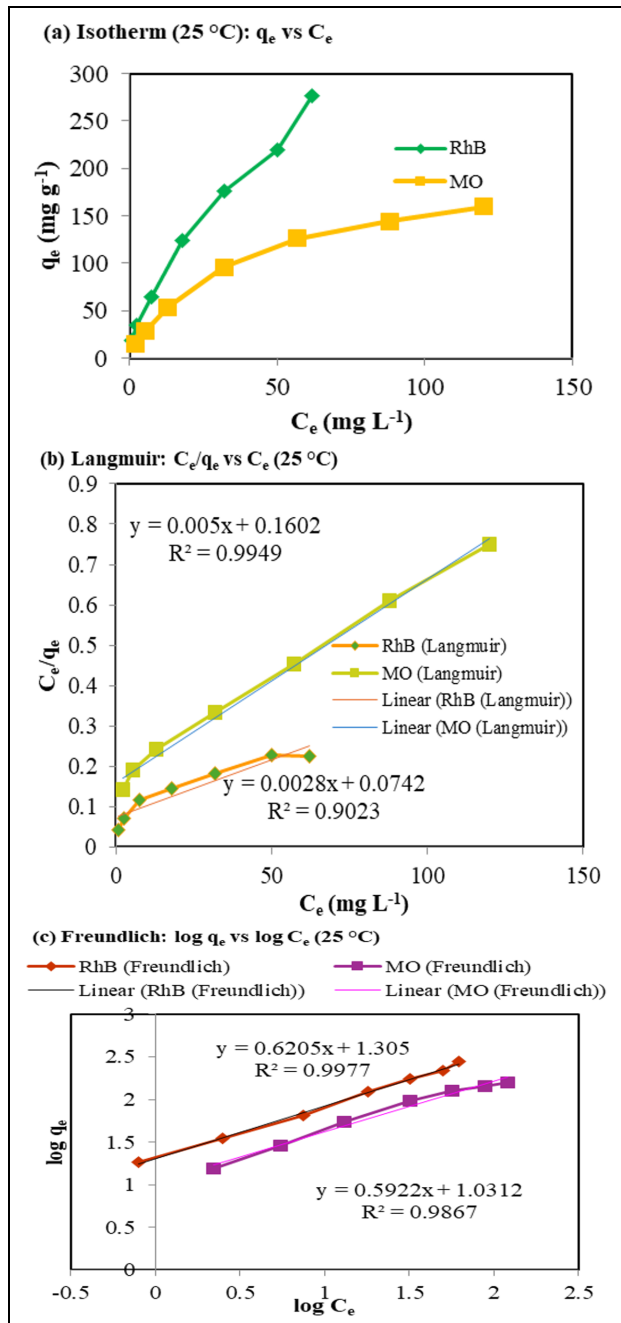


Fig. 9: Adsorption Isotherms and Isotherm Modeling of RhB and MO Dyes at 25 °C (a) Experimental adsorption isotherm (q_e vs C_e), (b) Langmuir isotherm model (C_e/q_e vs C_e), (c) Freundlich isotherm model ($\log q_e$ vs $\log C_e$)

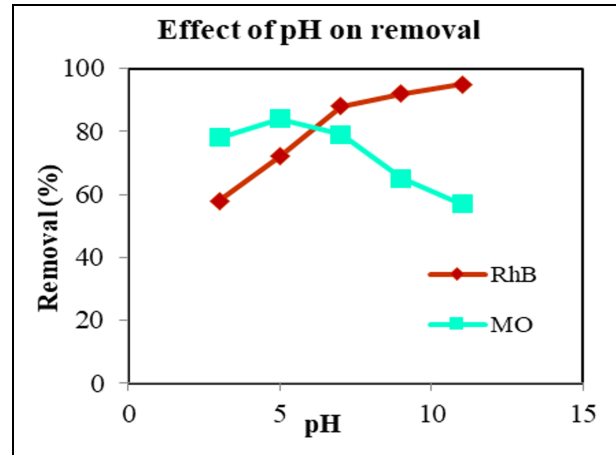


Fig. 10a: Effect of pH on the removal efficiency of RhB and MO Dyes

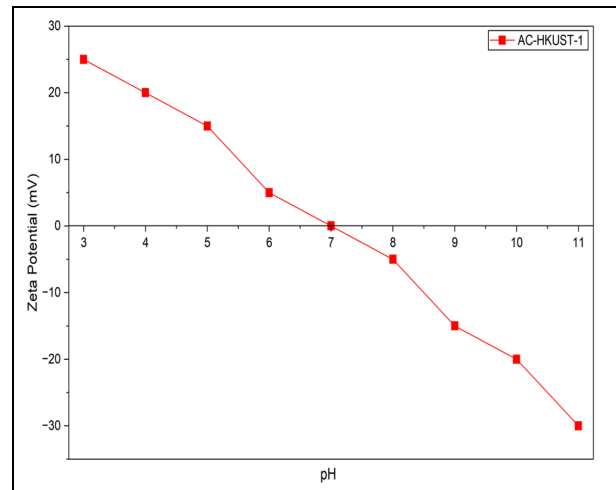


Fig. 10b: pH_{pzc} of the AC@HKUST-1 MOF

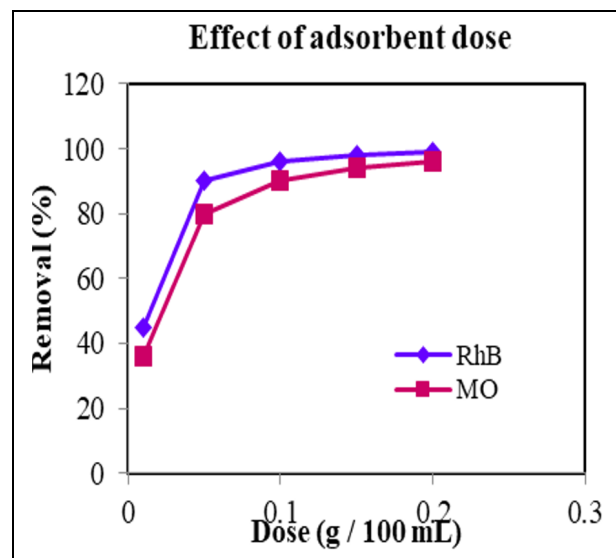


Fig. 11: Effect of adsorbent dose on removal efficiency of RhB and MO dyes

3.9 Effect of pH ($C_0 = 50 \text{ mg L}^{-1}$; 25°C)

For RhB, a cationic dye, sequestration increased gradually from 58% at pH 3 to 95% at pH 11 as shown in Fig. 10a. This trend can be attributed to the fact that at lower pH, the adsorbent surface is positively charged, causing repulsion with the positively charged RhB molecules, which results in lower adsorption. As the pH increases, the adsorbent surface becomes negatively charged, promoting strong electrostatic attraction with the positively charged RhB dye, thus enhancing its sequestration (Le *et al.* 2021). The pH_{PZC} of AC-HKUST-1 adsorbent was determined using the pH drift method and it was found to be 7 (Fig. 10b). Below this pH (i.e., when $\text{pH} < (\text{pH}_{\text{PZC}})$), the adsorbent surface is positively charged, which favors the adsorption of anionic dyes such as MO. Conversely, at pH values above pH_{PZC} (i.e., $\text{pH} > \text{pH}_{\text{PZC}}$), the adsorbent surface becomes negatively charged, facilitating the adsorption of cationic dyes like

RhB while repelling anionic dyes. These findings provide a clear explanation for the observed variations in the removal efficiencies of MO and RhB at different pH levels.

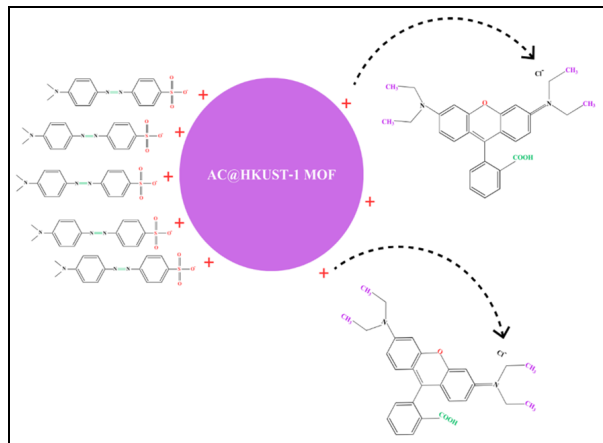


Fig. 12: Possible mechanism of RhB and MO AC@HKUST-1 MO

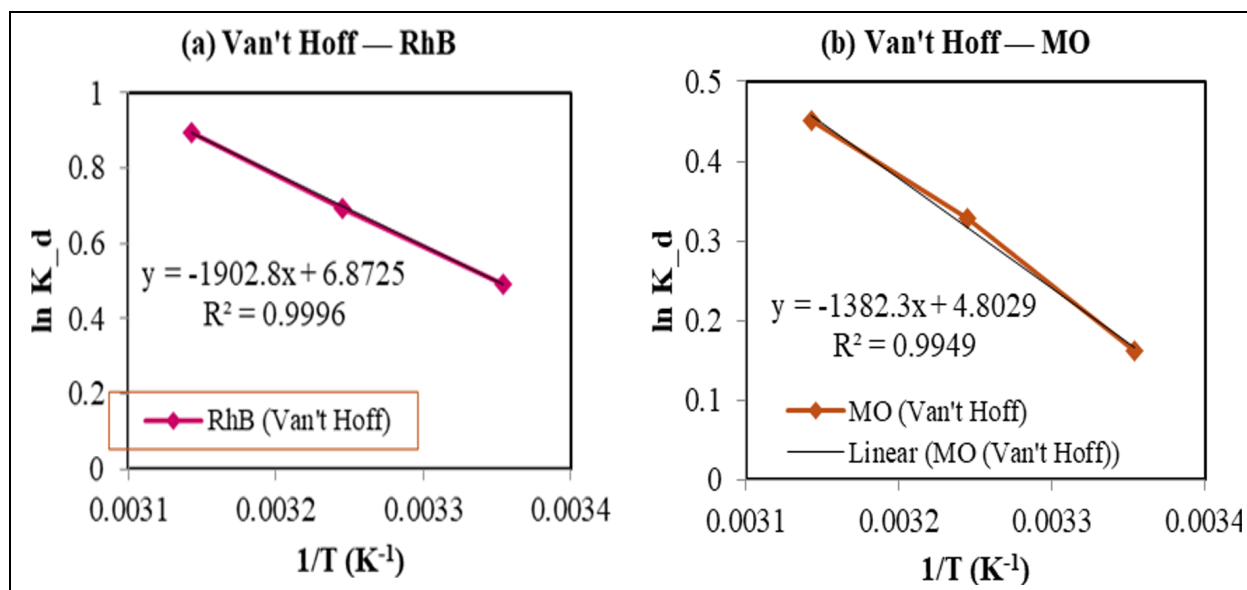


Fig. 13: (a) Van't Hoff plot for RhB adsorption and (b) Van't Hoff plot for MO adsorption

3.10 Effect of Adsorbent Dosage ($C_0 = 50 \text{ mg L}^{-1}$; pH 7; 25°C)

The impact of adsorbent concentration on dye removal was evaluated in the range of 0.01–0.20 g per 100 mL solution (Table 5; Fig. 11). For RhB, removal efficiency increased from 45% at 0.01 g to nearly 99% at 0.20 g, while MO removal improved from 36% to 96% under identical conditions. This improvement is attributed to the greater number of available active sites and enhanced probability of dye–adsorbent interactions

at higher dosages. However, the adsorption capacity expressed per unit mass declined with increasing dosage, a well-documented phenomenon in batch systems (Aziz *et al.* 2025). This reduction results from partial site overlap, aggregation of particles, and unsaturation of adsorption sites at high solid-to-liquid ratios. Comparable dosage-dependent behavior has been reported for other HKUST-1-derived and carbon-based composites, confirming the generality of this trend. Thus, 0.05–0.10 g appears to provide an optimal balance between removal efficiency and material utilization. Possible dye adsorption mechanism is shown in Fig. 12.

Table 7. Comparison of removal of MO and RB dyes by different adsorbents

MOF Adsorbent	Dye	Concentration of Dye (mg L ⁻¹)	q _{max} (mg g ⁻¹)	Time (min)	Reference
AC@HKUST-1 MOF	Rhodamine B	50	276	120	Current study
	Methyl orange	50	160	180	Current study
HKUST-1	Methyl orange	-	398.8	30	(Shen <i>et al.</i> 2018)
TiO ₂ @HKUST-1	Rhodamine B	-	95.2	60	(Qiao <i>et al.</i> 2023)
ZN(BDC) MOF	Aniline Blue	59.02	55.34	60	(Elsherbiny <i>et al.</i> 2023)
	Orange (II)	35.03	2.632	120	
	Methylene Blue	12	12	120	
MOF-5 COF (M5C)	Rhodamine B	16	16.18	8	(Firoozi <i>et al.</i> 2020)
MOF-5@GO	Rhodamine B	550 (ppm)	151.62	10	(Kumar and Masram 2021)
Fe-MOFs	Rhodamine B	18	70.75	300	(Wei <i>et al.</i> 2018)
Ni-MOF [Ni(II)L]	Rhodamine B	16	163.66	39	(Mohammadi <i>et al.</i> , 2023)
Fe-MOFs	Rhodamine B	50 (ppm)	135	240	(Ren <i>et al.</i> 2022)
DMACBUT-8(Cr)	Rhodamine B	175	318.6	150	(Nguyen <i>et al.</i> 2023)
H+CBUT-8(Cr)	Rhodamine B	175	811.7	150	(Nguyen <i>et al.</i> 2023)
MIL-68(Al)	Rhodamine B	5	1111.11	14	(Saghanejhad Tehrani and Zare-Dorabei 2016)
Ce-doped UiO	Rhodamine B	1200	754.4	24	(Dong <i>et al.</i> 2020)
UiO-66-NH	Rhodamine B	20	82.69 %	150	(Chen <i>et al.</i> 2015)
Zn-MOF	Rhodamine B	15	3.750	1440	(Zhang <i>et al.</i> 2018)
Ni doped FeO(OH)-NWs-AC	Rhodamine B	50	210.17	2	(Azad <i>et al.</i> 2016)
ZIF-8@PEI-CC	Methyl Orange	1200	917.43	360	(Lei <i>et al.</i> 2023)
ZIF-8	Methyl Orange	1200	326.79	120	(Lei <i>et al.</i> 2023)
MOF-2@Al ₂ O ₃	Methyl orange	250	300.3	5	(Koppula <i>et al.</i> 2022)
PCN-222 (MOF - 545)	Methyl orange	1000	154.1	50	(Li <i>et al.</i> 2017)
		600	128	50	
		400	76.54	50	
MOF - 235	Methyl Orange	1000 (ppm)	477	-	(Haque <i>et al.</i> 2011)
Ni@ZIF-6	Methyl Orange	20	17.69	180	(Nazir <i>et al.</i> 2023)
Zr-MOF	Methyl Orange	-	854.30	-	(Hu <i>et al.</i> 2024)
[Cd(INA)2(H ₂ O)].ISB.	Methyl Orange	200 (ppm)	166	-	(Tella <i>et al.</i> 2017)
Cu50-Cr50-MOF	Methyl Orange	100 (ppm)	271.7	90	(Salama <i>et al.</i> 2023)
Fe-BTC	Methyl Orange	15	100.3	120	(Karami <i>et al.</i> 2021)
ZIF-67/AC	Rhodamine B	120	46.2	60	(Li <i>et al.</i> 2017)
MIL-53-Fe	Rhodamine B	50	55	180	(Navarathna <i>et al.</i> 2020)

4. CONCLUSION

This study presents the synthesis of an activated carbon-HKUST-1 (AC@HKUST-1) nanocomposite, designed to enhance the adsorption and decontamination of both anionic and cationic dyes from wastewater. The composite was produced through an *in situ* ultrasonically assisted growth technique, combining the high surface area and stability of activated carbon with the porous structure of HKUST-1 MOF to achieve efficient dye removal. The composite demonstrated effective removal

kinetics and adsorption isotherms, presenting a promising solution for wastewater treatment. The findings highlight exceptional performance of the nanocomposite in removing RhB and MO, with a higher adsorption capacity for RhB. Additionally, AC@HKUST-1 could be a low-cost and environmentally friendly adsorbent for the effective removal of cationic pollutants from aqueous solutions. This makes it an attractive option for the development of affordable and sustainable adsorbents in water treatment applications. However, several important factors need to be considered

for its industrial applicability. The cost of the MOF precursor, specifically copper-based materials, could potentially limit the large-scale use of AC@HKUST-1. High precursor costs may make the material less economically viable in commercial operations, especially when compared to conventional adsorbents. Furthermore, the risk of leaching under extreme pH conditions remains a concern for long-term use in real-world applications. While the composite shows stability within the tested pH range, its performance under highly acidic or alkaline environments needs further investigation to prevent potential breakdown and contamination. To improve industrial competitiveness, the material's cost-effectiveness must be optimized without compromising its high adsorption performance. Future studies should aim to reduce production costs, enhance material stability under varying pH conditions, and explore potential modifications to improve the composite's resilience in complex industrial effluents.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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