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Adsorption and Isotherm Studies on Separation of Methyl Orange by Chitosan-modified activated carbon

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***Abstract*: Water contamination from industrial dyes such as Methyl Orange (MO) presents a significant environmental challenge due to their toxicity and persistence. In this study, low-cost activated carbon derived from Aloe Vera leaf waste was modified with chitosan to prepare a biodegradable adsorbent for MO removal. The removal of such carcinogenic, non-biodegradable, and environmentally harmful dyes from aqueous systems is critical to addressing sustainability concerns. The chitosan-modified activated carbon demonstrated enhanced adsorption capacity, achieving a maximum removal efficiency of 76.19% at pH 2, with equilibrium reached within 45 minutes. The adsorption process was well described by the Langmuir isotherm model, confirming monolayer adsorption on homogeneous surfaces. The use of aloe vera leaf waste, treated with chitosan, offers a low-cost, eco-friendly, and renewable solution. This biosorbent, rich in functional groups and derived from naturally abundant waste materials, shows considerable potential as a sustainable alternative for dye removal from wastewater.**

***Author Keywords*: Aloe Vera leaf waste, Adsorption, Green material, Methyl Orange, Chitosan-modified activated carbon.**

I. INTRODUCTION

Water contamination is one of the most critical environmental challenges of the twenty-first century. This problem is aggravated by the irregular discharge of wastewater containing various inorganic and organic pollutants, especially in developing countries (Sheth Y. et al., 2021; Buthale R. et al., 2018). Methyl Orange (MO), a widely used anionic dye that dissolves in water, is extensively employed in the printing, paper, textile, food, and pharmaceutical industries (Huang J. et al., 2010; Malkapuram S. et al., 2021). Due to its toxic nature and potential to cause allergic reactions upon skin contact, the removal of MO from aqueous solutions is of significant importance (Umpuch C. et al., 2013; Uppalwar et al., 2025).

To address this issue, several treatment methods have been developed, including photocatalysis, oxidation, biodegradation, adsorption, membrane filtration, ion exchange, photodecomposition, and electrolysis (Khaniabadi et al., 2016, Thakur and Sonawane 2019). Among these, adsorption stands out due to its operational simplicity, adaptability, high efficiency, and the absence of harmful byproducts. Furthermore, both the adsorbent and adsorbate can often be regenerated, enhancing the sustainability of the process.

Biochar-based materials have shown considerable promise as sustainable and effective adsorbents for removing heavy metals and organic contaminants, owing to their high surface area, porosity, and abundance of surface functional groups (Uppalwar et al., 2022; Gujar et al., 2022; Thakur A. et al., 2016). The conversion of locally available agricultural waste into activated carbon has received much attention due to its economic and environmental advantages. In addition to conventional sorbents, various alternative materials including chitosan, surfactant-modified montmorillonite, acrylic acid-grafted carbon, organo-clays, Fe_3O_4-activated carbon, lignin, graphite carbon nitride, pine sawdust, and metal-organic frameworks have been studied for their potential to remove MO from wastewater (Gujar et al 2012; Jadhav et al., 2024).

Tropical plants like *Aloe vera*, cultivated extensively in warm climates such as those in India and the United States, generate significant agro-waste during pharmaceutical and latex production. This waste, particularly aloe vera leaves, can be effectively utilized to produce activated carbon (Prajapati et al., 2020). Recent studies have explored sulfuric acid-modified activated carbon derived from leftover aloe vera leaves, which proved effective in adsorbing MO and aniline from synthetic

effluents. Process parameters such as contact time, pH, sorbent dosage, and initial pollutant concentration have been widely optimized to improve adsorption outcomes (Bethi B. et al., 2017; Gujar J. G. et al., 2023).

Chitosan, a renewable and biodegradable polymer, is particularly appealing due to its biocompatibility, non-toxicity, low cost, and abundant availability (Gujar J. G. et al., 2024). The incorporation of chitosan into biochar matrices enhances adsorption efficiency by combining chitosan's strong chemical affinity with biochar's structural advantages. It acts as a natural binder, increasing the retention of contaminants on the adsorbent surface (Vedula A. et al., 2021).

The textile industry is a major consumer of synthetic dyes, accounting for approximately 60% of global dye usage. During dyeing processes, around 10–15% of dyes are lost and 15–20% end up in wastewater, eventually entering natural water bodies (Wagh S. et al., 2012; Mahajan M. et al., 2024). Removing color from such industrial effluents poses a significant challenge, particularly for the pulp and paper, textile finishing, and dye manufacturing sectors (Gadhe A. et al., 2015; Raj W. et al., 2024). Physical treatment methods especially adsorption using porous supports have proven to be highly effective (Gavali A. et al., 2023). While biological treatments have been attempted, they often fall short due to the poor biodegradability of most dyes.

Although advanced chemical treatments such as photocatalytic degradation, ozonation, and chlorination offer better results than conventional methods, they often involve high operational costs and complex procedures. Other materials like coal, fly ash, red mud, and micellar-enhanced ultrafiltration systems have also been reported to remove dyes like methylene blue (Rahman M. et al., 2012; Jadhav et al., 2024). However, adsorption remains one of the most promising and scalable solutions for removing dye pollutants from aqueous systems (Megat et al., 2018; Sonawane S. et al., 2023).

Overall, wastewater treatment strategies such as adsorption, membrane separation, and redox-based methods are gaining prominence, with adsorption being particularly favored due to its efficiency, operational ease, affordability, and scalability (Khedkar et al 2013; Patil et al., 2022, Malika et al 2016).

Lignin exhibits a wide range of applications, particularly in the adsorption of impurities such as pesticides, pharmaceuticals, heavy metals, and dyes, making it highly suitable for water purification and reuse (Gujar J. et al., 2010). Studies have demonstrated that lignin can serve as an efficient and cost-effective alternative to synthetic polymers. In one such study, Methyl Orange (MO) was successfully removed from synthetic wastewater using activated carbon derived from leftover aloe vera leaves and subsequently modified with chitosan (Kingsley et al., 2023). The adsorption process was optimized by evaluating the influence of various parameters, including initial pollutant concentration, contact time, adsorbent dosage, and pH.

A range of physical and chemical techniques has been employed for dye removal from textile effluents, including membrane filtration, electrochemical oxidation, ozonation, advanced oxidation processes, and coagulation/flocculation methods (Kale P. et al., 2023; Sonawane S. et al., 2022). However, many of these techniques suffer from significant limitations, such as complex system designs, limited removal efficiency, and high operational and maintenance costs (Hussain S. et al., 2021).

In contrast, adsorption is widely recognized as an effective treatment method due to its high efficiency, operational simplicity, cost-effectiveness, and scalability from laboratory to industrial applications (Sonawane S. et al., 2021). The selection of an appropriate adsorbent significantly influences the performance of the adsorption process (Shimpi et al 2011; Liu Q. et al., 2015). A variety of materials including activated carbon, biochar, clays, natural substances, agricultural wastes, and synthetic materials along with some nanoparticles have been developed and used for dye removal (Thakur et al 2021, Thakur et al 2020; Dronkar M. et al., 2018; Gadhe A. et al., 2014).

Among these, biochar-based adsorbents have been extensively studied for their efficiency and environmental safety in removing a wide range of contaminants, owing to their high porosity, large surface area, and abundance of functional groups (Gautam K. et al., 2025; Kellner-Rogers et al., 2019, Malika et al 2023). However, the adsorption efficiency of unmodified biochar can be limited by its high pH, heterogeneous surface structure, and negatively charged surface.

To enhance performance, biochar can be modified by incorporating additives such as zeolites, metal oxides, and especially chitosan a biodegradable polymer rich in amino and hydroxyl functional groups (Malika M. et al., 2023). These groups facilitate stronger electrostatic interactions with metal ions and dye molecules. Chitosan-modified biochar synergistically combines biochar's high surface area with chitosan's high chemical affinity, thereby significantly improving adsorption capacity for pollutants such as phosphate, dyes, and heavy metals like Cd(II) and Pb(II) (Sonawane et al 2022; Crini G. et al., 2019, Kadam et al 2023, Malika et al 2025).

In conclusion, Methyl Orange and other industrial dyes represent a major source of water contamination and pose a severe threat to aquatic ecosystems. Among the various treatment methods explored, biochar-based materials modified with chitosan offer a highly efficient, easy-to-use, and environmentally sustainable solution for dye removal from wastewater (Malika and Sonawane, 2022; Katubi K. et al., 2021; Sonawane S. et al., 2022, Kadam et al 2025).

This study employs chitosan-modified activated carbon synthesized from leftover *Aloe vera* leaves as a cost-effective and environmentally sustainable adsorbent for the removal of Methyl Orange (MO) dye from aqueous solutions. The focus is on valorizing *Aloe vera* leaf waste a widely available agricultural byproduct to produce activated carbon, which is

subsequently functionalized with chitosan to enhance adsorption performance. This dual strategy not only leverages low-cost, biodegradable, and renewable materials, but also significantly improves adsorption efficiency by introducing additional functional groups from chitosan.

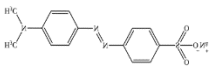
The work presents a novel integration of waste valorization and environmental remediation, offering a green and sustainable alternative to conventional adsorbents. A systematic comparison between untreated and chitosan-treated adsorbents is carried out under varied experimental conditions to evaluate improvements in performance and reusability. The findings highlight the potential of the developed adsorbent as an economical and scalable solution for industrial wastewater treatment, with broader applicability to other pollutants as well.

II. MATERIALS AND METHODS

Aloe vera waste was collected from households using aloe vera plants. The chemical reagents used in this study acetic acid, methyl orange (MO), sodium hydroxide (NaOH), sulfuric acid (H_2SO_4), and chitosan were procured from certified chemical suppliers. **Table 1** presents the physicochemical properties and molecular structure of methyl orange.

A digital pH meter was used to adjust the solution pH as required, using dilute solutions of NaOH and H_2SO_4. A stock solution of methyl orange (100 mg/L) was prepared in distilled water, and working solutions of desired concentrations were obtained by serial dilution.

TABLE 1
The chemical structure and general properties of Methyl orange

Generic name	Chemical formula	Chemical structure	Mm (g/mol)	λmax (nm)
Methyl orange	$C_{14}H_{14}N_3NaO_3S$		327.23	415

Aloe Vera

Fig. 1.- Dried aloe vera leaves waste powder

For centuries, *Aloe vera* has been highly regarded for its diverse medicinal applications, including the treatment of burns, wound healing, and potential roles in the prevention of chronic diseases such as diabetes and cancer. The gel extracted from the plant's leaves must be processed with care to preserve its bioactive compounds, which are responsible for its therapeutic properties. Ensuring product safety and efficacy, especially for pharmaceutical and nutraceutical applications, requires strict regulatory oversight. Historically, *Aloe vera* was revered across ancient civilizations from Egypt to China not only as a healing plant but also as a symbol of health and longevity. It was often referred to as the "plant of immortality" by the Egyptians and the "elixir of youth" in Chinese tradition. The name *Aloe* is believed to originate from the Arabic word "Alloeh", meaning "shining bitter substance," reflecting both its medicinal potency and historical significance.

Preparation of adsorbent

Fig.2.- Modified activated carbon-based Aloe vera leaves waste

After separating the gel from the *Aloe vera* leaves, the remaining fibrous waste was thoroughly washed with distilled water to eliminate residual impurities. The cleaned biomass was then shade dried for 4–5 days, followed by oven-drying at 150 °C for 5 hours to ensure complete moisture removal.

The dried *Aloe vera* leaf waste was ground using a laboratory mill and sieved to obtain particles of approximately 40 mesh size. These particles were then subjected to carbonization in a muffle furnace at 550 °C for 20 minutes to produce biochar.

Post-carbonization, the biochar was immersed in a 0.1 N H_2SO_4 acid solution and soaked for 12 hours to enhance surface activation. The treated sample was then filtered and repeatedly rinsed with distilled water until neutral pH was achieved, ensuring removal of residual chemicals.

Finally, the activated carbon was dried in an electric oven at 105 °C for 6 hours. The resulting material modified *Aloe vera*-based activated carbon was used as the adsorbent for all subsequent adsorption experiments (Fig. 2).

Treatment with chitosan:

One gram of dried powdered chitosan was dissolved in 0.05 L of 2% aqueous acetic acid, and it was then mixed with one gram of chitosan dissolved in 0.05 L of 1 M NaOH to form a chitosan-modified adsorbent. Five grams of modified activated carbon derived from discarded aloe vera leaves were mixed with this combination. After three hours of vigorous stirring at 60 °C, the mixture was poured onto a porcelain dish and left to dry at room temperature. To eliminate air bubbles, the mixture was kept at room temperature for 0.5 hours. After that, the modified adsorbent was crushed, and

sieved to 100 mesh, and its elemental composition was determined using Table 2 (Khan et al, 2019; Ni Z. et al.,2007).

TABLE 2
Elemental analysis of the chitosan membrane

	C	N	O
Chitosan	24.72	54.03	21.24

Batch adsorption experiments

Batch adsorption experiments were conducted to investigate the effects of key process variables, including solution pH (2–10), adsorbent dosage (2–5 g), initial methyl orange (MO) concentration (20–100 mg/L), and contact time (1–90 minutes). For each experiment, 100 mL of MO solution was placed in a 250 mL cylindrical glass beaker and mixed with a predetermined amount of chitosan-modified activated carbon adsorbent.

The mixtures were agitated at a constant speed of 200 rpm using a magnetic stirrer at ambient temperature (25 ± 2 °C) for the specified duration. Following adsorption, the solutions were filtered using Whatman No. 6 filter paper to remove suspended adsorbent particles. The residual MO concentration in the filtrate was measured using a UV–Visible spectrophotometer.

All experiments were performed in duplicate, and the average values were used for data analysis to ensure reproducibility and accuracy. The adsorption capacity of the sorbent was calculated using the following equation:1

$$qe = \frac{(Co-Ce)V}{m} \quad \text{.....(1)}$$

Where qe (mg/g) is the equilibrium absorption capacity of the adsorbates per gram of adsorbent. The contaminants' initial and equilibrium concentrations were denoted by the parameters C_o and C_f (mg/l), correspondingly. In addition, the volume of the solution is V (l), and the mass of the adsorbent is m (g). Eq. (2) was utilized to determine the percentage of dye removal.

$$\% \text{ Removal} = \left[\frac{Co-Cf}{Co}\right] \text{ x } 100 \quad \text{.....(2)}$$

Where C_f is the final concentration of dye after adsorption, C_0 is the initial concentration of dye before adsorption.

Methods of Analysis

The adsorption of methyl orange (MO) from aqueous solutions was carried out using chitosan-modified activated carbon prepared from *Aloe vera* leaf waste. The presence of functional groups responsible for MO binding such as amino (–NH_2) and hydroxyl (–OH) groups was confirmed through Fourier Transform Infrared Spectroscopy (FTIR), indicating successful surface modification of the adsorbent.

To assess removal efficiency, MO concentrations were determined using UV–Visible spectroscopy by measuring the absorbance at the dye's maximum wavelength (λmax) before and after adsorption.

The adsorption behavior was modeled using the Langmuir isotherm, which assumes uniform monolayer adsorption on a homogenous adsorbent surface. This model was applied to evaluate and optimize key parameters influencing the adsorption process, including contact time, adsorbent dosage, solution pH, and initial dye concentration.

III. RESULTS AND DISCUSSION

Characterization

Figure 3(a) and 3(b) present the FTIR spectra of activated carbon derived from *Aloe vera* leaf waste, both before and after the adsorption of methyl orange (MO). Prior to adsorption, characteristic absorption bands were observed at 478, 622, and 1156 cm^{-1}, corresponding to Si–O–Si symmetric stretching, Si–O–M (where M = Al or Mg) bending vibrations, and Si–O stretching vibrations, respectively indicating the presence of silicate structures on the adsorbent surface.

After adsorption, a broad peak appeared in the range of 3416–3552 cm^{-1}, attributed to O–H stretching vibrations. This shift suggests increased hydrogen bonding and vibrational interactions of water molecules, likely induced by the binding of MO and associated contaminants onto the surface of the activated carbon. These spectral changes confirm successful interaction between the dye molecules and functional groups present on the modified adsorbent surface.

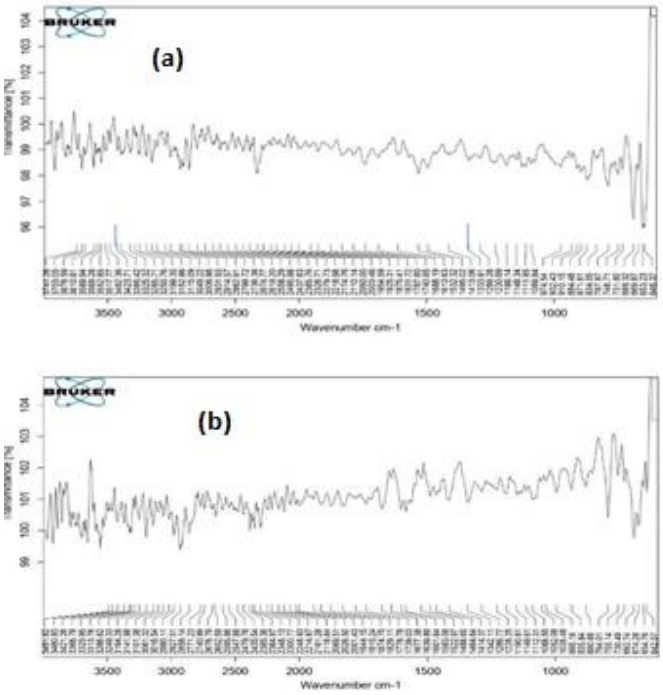


Fig. 3.- FTIR Spectra of activated carbon-based Aloe Vera leaves waste (a) before & (b) after the adsorption.

Figure 4 (a–b) illustrates the FTIR spectra of chitosan,

revealing the presence of characteristic functional groups. A broad band in the range of 3291–3361 cm^{-1} corresponds to overlapping N–H and O–H stretching vibrations, along with intramolecular hydrogen bonding, indicative of chitosan's highly hydrophilic nature.

Distinct absorption bands at 2921 cm^{-1} (C–H symmetric stretching) and 2877 cm^{-1} (C–H asymmetric stretching) are characteristic of the polysaccharide backbone. The presence of N-acetyl groups was confirmed by peaks at 1645 cm^{-1}(C=O stretching of amide I) and 1325 cm^{-1} (C–N stretching of amide III). A weak band near 1550 cm^{-1}, corresponding to N–H bending of amide II, may have overlapped with nearby absorptions.

Additional bands at 1423 cm^{-1} and 1375 cm^{-1} correspond to CH_2 and CH_3 bending vibrations, respectively, while a distinct band at 1153 cm^{-1} suggests C–O–C bridge stretching, typical of glycosidic linkages. Finally, C–O stretching vibrations were observed at 1066 cm^{-1} and 1028 cm^{-1}.

These spectral features are consistent with the well-established chemical structure of chitosan, confirming its purity and functional integrity for use as a surface-modifying agent in the adsorbent system.

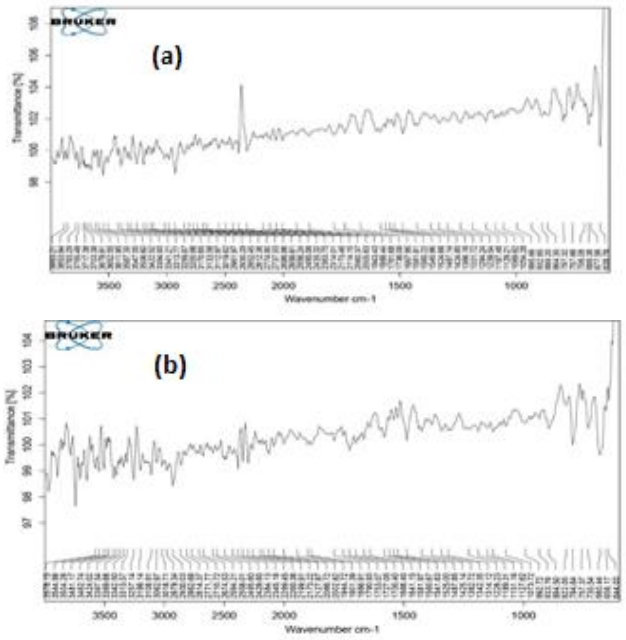


Fig. 4- FTIR Spectra of modified activated carbon-based Aloe Vera leaves waste (a) before & (b) after the adsorption

Effect of various parameters

The adsorption process is influenced by several key parameters that determine the efficiency and capacity of adsorbent removal from solutions. The factors that are evaluated include contact time, pH, concentration and adsorbent dose.

Effect of contact time Untreated/Treated adsorbent

The effect of contact time and initial dye concentration on the adsorption of methyl orange (MO) was evaluated using both untreated and chitosan-modified *Aloe vera*-based activated carbon. For the untreated adsorbent, MO uptake increased gradually over time, reaching adsorption equilibrium at approximately 60–70 minutes. The maximum removal efficiency was 50.84% at an initial dye concentration of 4 ppm, which increased to 69.62% at 12 ppm, indicating limited adsorption capacity and slower kinetics at higher concentrations.

In contrast, the chitosan-modified adsorbent exhibited significantly faster adsorption kinetics, attaining equilibrium within 35–45 minutes. It also demonstrated markedly higher removal efficiencies, with 70.00% at 4 ppm, 67.44% at 6 ppm, and 63.10% at 12 ppm. These results clearly indicate that chitosan functionalization enhances both the adsorption rate and overall efficiency, particularly under higher contaminant loads.

This improvement can be attributed to the increased number of functional binding sites (e.g., $–NH_2$, –OH groups) introduced by chitosan, which facilitate stronger electrostatic interactions and hydrogen bonding with the anionic MO dye molecules.

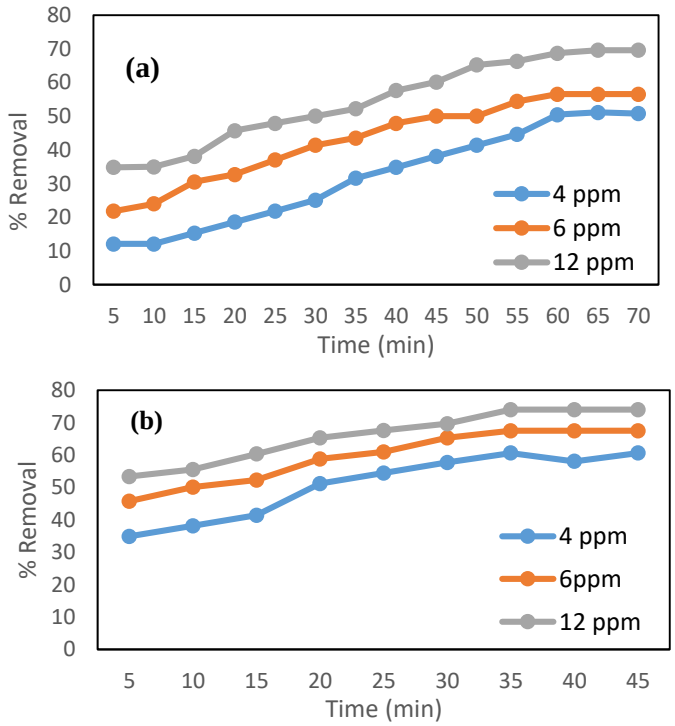


Fig.5. Effect of contact time of adsorbent on % MO removal (a) untreated (b) treated

Effect of pH

The effect of solution pH on methyl orange (MO) adsorption was evaluated for both untreated and chitosan-modified *Aloe vera*-based activated carbon. For the untreated adsorbent, removal efficiency decreased with increasing pH,

with a maximum adsorption of 54.42% observed at pH 2. This trend is attributed to enhanced protonation of the adsorbent surface under acidic conditions, resulting in a more positively charged surface that favors electrostatic attraction with the anionic MO dye.

Similarly, the chitosan-modified adsorbent showed reduced adsorption at higher pH levels, but achieved a significantly higher maximum removal efficiency of 73.95% at pH 2. This enhanced performance is due to the increased surface protonation of chitosan under acidic conditions, which amplifies electrostatic interactions between the adsorbent and the negatively charged dye molecules.

Based on these findings, pH 2 was identified as the optimal condition for MO removal using both adsorbents. The treated adsorbent consistently outperformed its untreated counterpart, highlighting the beneficial role of chitosan in improving adsorption efficiency under low pH environments.

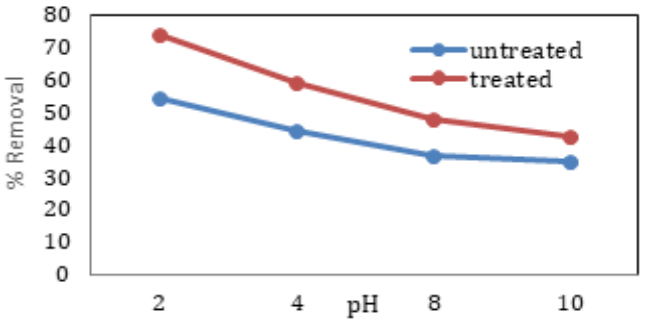


Fig. 6.- Effect of pH on percentage removal of MO dye on a chitosan-modified Aloe vera waste

Effect of adsorbent dosage Untreated/ Treated Adsorbent

Figure 7 illustrates the effect of adsorbent dosage (ranging from 0.5 to 2.5 g/L) on the methyl orange (MO) removal efficiency using both untreated and chitosan-modified *Aloe vera*-based activated carbon. For the untreated adsorbent, a gradual increase in MO adsorption capacity was observed as the dosage increased, rising from 39.23 mg/g at 0.5 g/L to 61.80 mg/g at 2.5 g/L. This increase is attributed to the greater availability of active adsorption sites at higher dosages.

In comparison, the chitosan-modified adsorbent exhibited significantly higher adsorption performance across the same dosage range. The adsorption capacity increased from 65.27 mg/g at 0.5 g/L to 71.35 mg/g at 2.5 g/L, indicating enhanced interaction between the dye molecules and the modified adsorbent surface.

These results confirm that chitosan functionalization enhances the adsorbent's efficiency, even at lower dosages, by introducing additional functional groups that promote dye binding. Thus, the chitosan-modified *Aloe vera* adsorbent proves to be more effective and economical for MO removal.

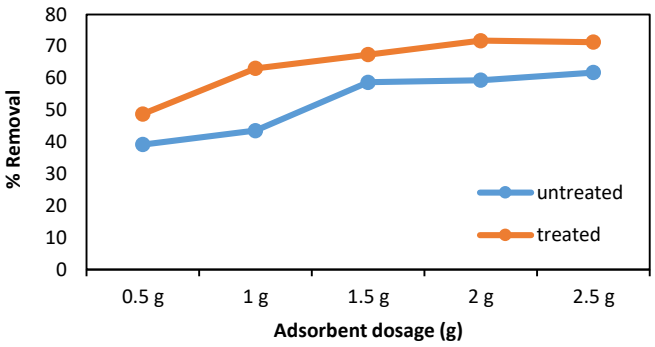


Fig. 7.- Effect of adsorbent dose on percentage removal of MO dye on a chitosan-modified Aloe vera waste

Effect of initial adsorbate concentration

Figure 8 illustrates the influence of initial methyl orange (MO) concentration on its adsorption by both untreated and chitosan-modified *Aloe vera*-based activated carbon. The experiments were conducted at room temperature (25 °C) using a fixed adsorbent dosage of 1 g/L, with contact times ranging from 5 to 50 minutes. For the untreated adsorbent, MO removal efficiency increased significantly as the initial dye concentration increased from 4 mg/L to 12 mg/L, with removal efficiencies improving from 25.78% to 66.98%. This enhancement is attributed to increased molecular interactions, such as Van der Waals forces, at higher dye concentrations, which drive greater adsorption onto the available active sites. In the case of the chitosan-modified adsorbent, a consistent and more pronounced increase in removal efficiency was observed, rising from 38.15% at 4 mg/L to 72.28% at 12 mg/L. This superior performance can be credited to the enhanced surface functionality imparted by chitosan, which introduces additional binding sites (e.g., $–NH_2$ and –OH groups) and promotes stronger electrostatic and hydrogen-bonding interactions with the anionic dye molecules.

Overall, these findings confirm that chitosan modification significantly improves the adsorption capacity of the bio-based adsorbent, particularly under higher pollutant loading conditions.

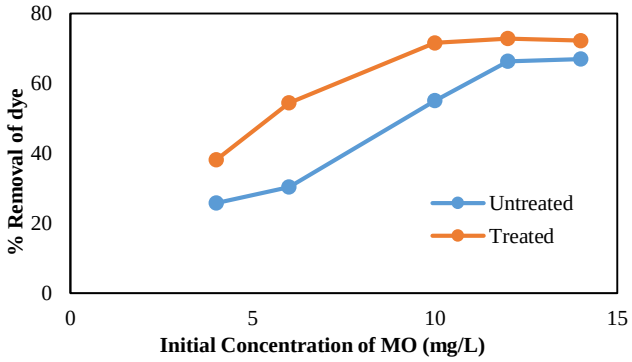


Fig. 8.- Effect of initial adsorbate concentration on percentage removal of MO dye on a chitosan-modified Aloe vera waste

Adsorption, kinetics and isotherms

Equation (3) was utilized to obtain the dye removal.

$$\% \, Removal = \left[\frac{C_0 - C_t}{C_o}\right] \quad \text{.....(3)}$$

Where C_0 represents the dye's initial concentration before adsorption. C_t is the dye's concentration following adsorption. Using Equation, the amount of adsorbed dye at equilibrium was determined (4).

$$qe = \frac{(Co - C_t)V}{m} \quad \text{..... (4)}$$

where C_t is dye concentration (ppm) during various time periods Adsorbent weight (g) is denoted by w, solution volume (ml) by v, and starting concentration (ppm) by C_0. The adsorption isotherm was used to determine the link between the mass and the concentration of adsorbed dye. The adsorption isotherm was computed using the experimental data (Huang R. et al., 2014).

Kinetic modelling

A given time t and the quantity of dye adsorbed at equilibrium are denoted by q_e and q_t, respectively, in this straightforward pseudo-first order equation adsorption kinetic model. The form of this on integration in time t is linear and is

$$\frac{dq_t}{d_t} = k_1 (q_e - q_t) \quad \text{.....(5)}$$

$$log(q_e - q_t) = ln\ (q_e) - k_1\, t \quad \text{.....(6)}$$

To evaluate the kinetic behavior of methyl orange (MO) adsorption onto both untreated and chitosan-modified *Aloe vera*-based activated carbon, the experimental data were analyzed using pseudo-first-order and pseudo-second-order kinetic models.

Pseudo-First-Order Model

A plot of time (t) versus log (qe – qt) was constructed to examine the applicability of the pseudo-first-order kinetic model. The rate constant (k_1) was obtained from the slope or intercept of the linear fit to the experimental data. As shown in Figure 9(a)**,** the value of k_1 decreased with increasing dye concentration, which can be attributed to intensified competition for the limited number of active adsorption sites. This initial drop in the rate constant implies that lower initial dye concentrations (C_0) favor more efficient uptake in the early stages of adsorption. The observed trend suggests that the adsorption mechanism at lower concentrations is primarily governed by film diffusion, where the bulk dye molecules are rapidly transported to the adsorbent surface. This aligns with findings reported by (Nourmoradi H. et al., 2012), where diffusion dominates the mass transfer process in early adsorption stages.

Pseudo-Second-Order Model

To assess the pseudo-second-order kinetics, a plot of time (t) versus t/qt was generated. The linear regression fit provided the rate constant (k_2) from the slope and intercept of the plot. The pseudo-second-order model assumes that chemisorption is the rate-limiting step involving valence forces through electron sharing or exchange between adsorbent and adsorbate.

Figure 9(b) depicts the fitting of this model. As dye concentration increased, a decrease in k_2 was observed, again indicating competitive interactions for limited active sites, which can suppress adsorption rates at higher C_0 values. The intercept of the curve provides insight into the thickness of the boundary layer, reflecting resistance to mass transfer at the adsorbent surface (Haddadi E. et al., 2023; Parate et al 2011).

Overall, the data suggest that the pseudo-second-order model better describes the adsorption process due to higher correlation coefficients and closer agreement between calculated and experimental q_e values. The underlying mechanism appears to be controlled by diffusion coupled with surface interaction, particularly in the case of the chitosan-modified adsorbent.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}\, t \quad \text{.....(7)}$$

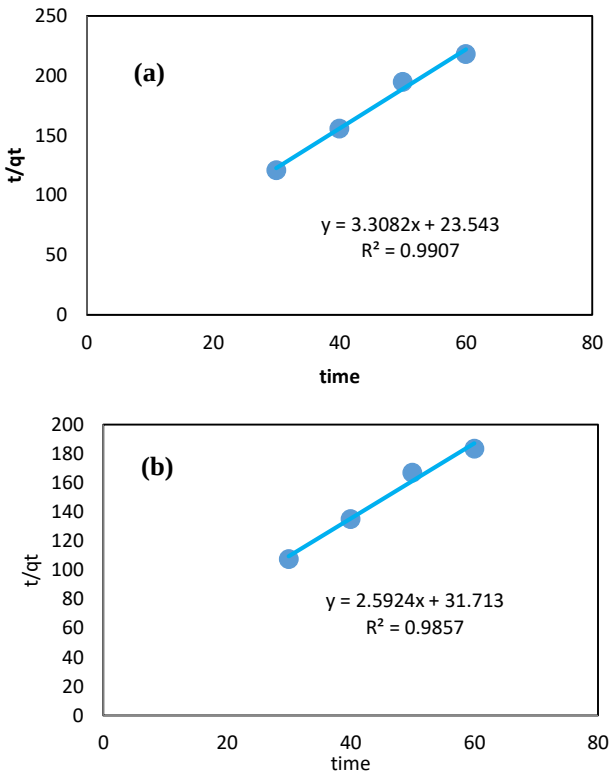


Fig. 9.- Pseudo-second order (a) Untreated (b) Treated

Langmuir adsorption isotherm

The combined structural and functional behavior of the adsorbent and the adsorbate determines the utilized adsorbent's

adsorption capability. Because of its planar structure, methyl orange may easily be adsorbed on membranes via van der Waals force and H-bonding interaction. The Langmuir model, a well-known model for monolayer adsorption with a minimal number of adsorption sites and homogeneous adsorption energies, was used to evaluate and assess the membranes' adsorption capacity as follows :

$$\frac{1}{q_e} = \left(\frac{1}{k_a q_m}\right)\frac{1}{c_e} + \frac{1}{q_m} \quad \ldots\ldots\ldots\ldots\ldots\ldots (8)$$

The separation factor, or dimensionless constant, is derived from the following and is a crucial property of the Langmuir isotherm.

$$R_L = \frac{1}{1+k_L C_o} \quad \ldots\ldots\ldots\ldots\ldots\ldots (9)$$

The adsorption is denoted by R_L values as irreversible when R_L = 0, linear when R_L = 1, favorable when 0<1, and unfavorable when R_L > 1. R_L in this study ranges from 0 to 1 (Table 2). An idea of the Langmuir adsorption at a given adsorbent weight is provided in Fig. 10 (a) & (b) (Huang R. et al., 2014).

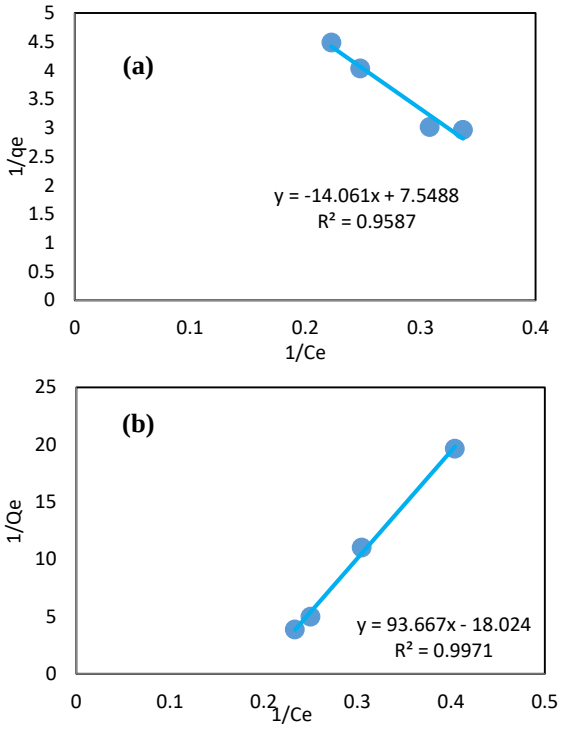


Fig. 10.- Langmuir adsorption isotherm for adsorption of Methyl Orange (a) Untreated (b) Treated

Freundlich adsorption isotherm

This isotherm describes reversible adsorption rather than a monolayer formation. where the Freundlich constants are k_f and n. The Freundlich isotherm constants k_f and n, which are related to adsorption intensity and capacity, respectively, are plotted against log Ce in the graph. The following is the equation:

$$log\ q_e = log\ K_F + \frac{1}{n}\ log\ C_e \quad \ldots\ldots\ldots\ldots\ldots\ldots (10)$$

The intercept and slope of the plot provide the values. Since the process is better matched by the Langmuir isotherm (0.99) than by the Freundlich model, the Langmuir values are compared with the Freundlich parameters in Table 2. Fig. 11 (a) & (b) shows the Freundlich adsorption at a certain adsorbent weight. The correlation coefficient, or R^2, indicates the strength of fit. The q_m value, which was ascertained using the Langmuir isotherm, and the anticipated removal efficiency matched rather well. The separation factor (R_L) of the Langmuir model, which ranges from 0 to 1, would suggest that MO will adsorb well and that the adsorption procedure is doable. This proves that adsorption in monolayers happens spontaneously (Nourmoradi H. et al., 2012; Hakke et al 2021).

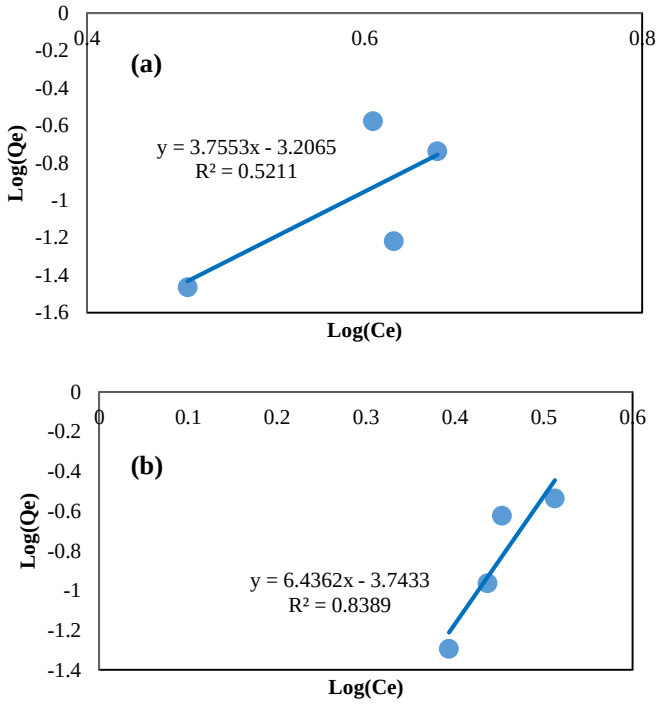


Fig. 11.- Freundlich adsorption isotherm for adsorption of Methyl Orange (a) Untreated (b) Treated

Temkin Isotherm

This isotherm includes a factor that specifically accounts for the interactions between the adsorbent and the adsorbate. The model implies that the heat of adsorption (function of temperature) of all molecules in the layer will decrease linearly rather than logarithmically with coverage, as equation (Kellner-Rogers et al., 2019) indicates, by ignoring the extremely low and large concentration values. As shown in Fig. 12 (a) & (b), a consistent distribution of binding energies (up to a maximum binding energy) was possible by plotting sorbed qe versus ln C_e. The constants were discovered by examining the slope and intercept. The model can be generated using the equation (11).

$$q_e = \frac{R\,T}{b}\, ln\, A_T + \frac{R\,T}{b}\, ln\, C_e \quad \ldots\ldots\ldots\ldots\ldots\ldots (11)$$

Langmuir and Freundlich kinetics parameters for MO dye removal have been provided in Table 2.

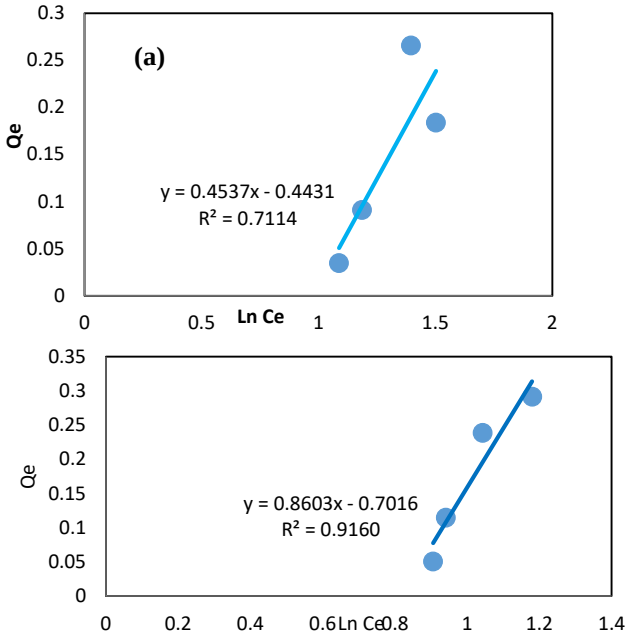


Fig.12.- Temkin adsorption isotherm for adsorption of Methyl Orange (a) Untreated (b) Treated

TABLE 3
Langmuir, Freundlich, Temkin kinetics parameters for Methyl Orange dye removal

Studies	Model	Treatment	R^2
Kinetic Model	Pseudo Second Order	Untreated	0.9907
		Treated	0.9857
Isotherm	Langmuir	Untreated	0.9587
		Treated	0.9971
	Freundlich	Untreated	0.5211
		Treated	0.8389
	Temkin	Untreated	0.7114
		Treated	0.9160

Adsorption data indicates that both untreated and chitosan-treated adsorbents exhibit pseudo-first-order kinetics, with high R^2 values shows in Table 3. Pseudo First Order kinetics demonstrated a strong connection between untreated (R^2 = 0.9907) and treated (R^2 = 0.9857) adsorbents. This shows that MO adsorption on both types of adsorbents has pseudo-first-order kinetics, which means that the adsorption rate is most likely proportional to the number of vacant active sites. The Langmuir isotherm best matches the results, notably for the treated adsorbent (R^2 = 0.9971), suggesting improved monolayer adsorption (Wu G. et al., 2012). The Freundlich and Temkin models fit better following chitosan treatment, indicating increased surface heterogeneity and greater adsorbate-adsorbent interactions. Overall, chitosan modification enhances adsorption efficiency and model fitting.

Key Issues and Challenges

Despite significant advancements in adsorbent development and performance optimization, the real-world deployment of adsorption technologies for dye separation continues to face multifaceted challenges. Scalability, cost-effectiveness, adsorbent regeneration, and adaptability to diverse industrial effluents remain major constraints limiting the transition of laboratory-scale research to industrial-scale applications.

Material Limitations and Selectivity:

While the chitosan-modified activated carbon derived from Aloe vera waste demonstrated promising removal efficiency for Methyl Orange (MO), a critical issue persists—limited selectivity across a wide spectrum of dye classes and complex wastewater matrices. Most adsorbents, including biochar-based materials, exhibit high affinity only for specific contaminants, limiting their universal applicability. Furthermore, real effluents contain mixed pollutants that may interfere with the adsorption process through competitive interactions.

Sustainability and Regeneration:

Although bio-based materials like chitosan and agro-waste-derived carbon offer sustainable alternatives, maintaining adsorption efficiency over multiple regeneration cycles remains a technical hurdle. Thermal and chemical regeneration techniques can degrade material properties or require high energy inputs, thereby reducing the overall sustainability and cost-efficiency of the process.

Process Optimization and Variability in Operating Conditions:

Adsorption performance is highly sensitive to operating parameters such as pH, contact time, initial dye concentration, and adsorbent dosage as evidenced in the present study. Maintaining these optimized conditions consistently in a dynamic industrial setting is challenging. The need for adaptive systems that can respond to real-time fluctuations in effluent composition is critical for practical deployment.

Data-Driven Material Design and AI Integration:

The advent of artificial intelligence (AI) and machine learning (ML) provides transformative tools for predictive modeling, adsorbent design, and process control. These technologies can facilitate the forecasting of adsorption behavior under varying environmental and compositional scenarios, and support the rational design of multifunctional adsorbents with tunable surface chemistry. AI-assisted frameworks can also accelerate kinetic and isotherm model fitting, as demonstrated in recent studies, enabling rapid screening and scale-up of adsorbents.

Instrumentation and Characterization Advances:

High-resolution analytical techniques such as Fourier-transform infrared spectroscopy (FTIR), UV-Vis spectroscopy, scanning electron microscopy (SEM), and high-performance liquid chromatography (HPLC) are now indispensable for unraveling the physicochemical mechanisms of adsorption. These methods offer deeper insights into surface functionalities, interaction mechanisms, and structural evolution during adsorption and regeneration cycles. However, the need for integrating these techniques into a continuous monitoring framework for industrial application is still unmet.

Regulatory and Environmental Compliance:

Lastly, achieving compliance with effluent discharge standards while ensuring economic feasibility remains a pressing concern. Adsorbents must not only meet performance metrics but also satisfy biodegradability, non-toxicity, and post-treatment disposal criteria.

IV. CONCLUSION

The present study demonstrates that chitosan-modified activated carbon derived from *Aloe vera* leaf waste serves as an efficient, low-cost, and eco-friendly adsorbent for the removal of methyl orange (MO) from aqueous solutions. Key parameters influencing adsorption such as contact time, pH, adsorbent dosage, and initial dye concentration were systematically evaluated. Equilibrium was achieved within 60 minutes, with maximum MO removal occurring at pH 2, highlighting the critical role of acidity in enhancing adsorption efficiency. The adsorption data were best described by the Langmuir isotherm model, indicating monolayer adsorption on a homogenous surface. A maximum removal efficiency of 76.19% was observed using the chitosan-modified adsorbent, confirming its superior performance.

The results affirm the potential of this natural, nontoxic, and renewable adsorbent prepared from agricultural waste and biopolymer chitosan as a promising material for dye removal. Its high adsorption capacity, environmental sustainability, and economic viability position it as a suitable alternative to conventional synthetic adsorbents for wastewater treatment applications.

Conflicts of Interest

The author(s) declared no conflicts of interest concerning the research, authorship, and/or publication of this article.

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