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Sustainable Dye Removal A Review of Low-Cost Natural Adsorbents for Wastewater Treatment

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ABSTRACT
Background: *Wastewater discharge into natural watercourses causes pollution of surface and groundwater, making it unsuitable for drinking and increasing industrial treatment costs. Color is a major pollutant, as even small amounts of dyes make water visually unappealing. Adsorption is a widely used physicochemical treatment method, where wastewater is mixed with an adsorbent to remove contaminants.*
Materials and Methods: *this study used natural adsorbents—Moringa seed and lemon seed powders—to remove methylene blue dye from synthetic textile wastewater. The seeds were sourced from local markets and farms, dried, and sieved. Adsorption experiments were conducted by varying contact time, initial dye concentration, pH, and adsorbent dosage.*
Results: *Lemon seed powder achieved 86% color removal at pH 7 with a 30-minute equilibrium time, while Moringa seed powder achieved 50% removal at pH 6 with a 60-minute equilibrium time. Experimental data fit both Langmuir and Freundlich adsorption isotherms.*
Conclusion: *Lemon seed powder proved significantly more effective than Moringa seed powder for methylene blue removal. Both materials show potential as low-cost, eco-friendly adsorbents that can reduce wastewater treatment expenses.*
Keywords: *Textiles Wastewater, synthetic wastewater, methylene blue dye, adsorption, natural adsorbents, Moringa seeds, lemon seeds, adsorption isotherms.*

INTRODUCTION
Textiles, Rubber, Plastics, Printing, Leather, Cosmetics, Ink, and other sectors frequently utilize dyes to color their goods. They consequently produce an enormous volume of colorful wastewater. Over 7 X 105 tonnes of dye are generated annually, and there are over 10,000 commercially available dyes. According to estimates, related sectors release 2% of the dyes produced each year as effluents (Easton, 1995). The textile sector uses the most dyes to color fibers among all other industries. One of India's oldest industries is the textile sector. In terms of employment in the nation, it is merely the second-largest industry after agriculture. With the second-highest population in the world, this growing nation has enormous potential for rapid industrial growth. One of these is the textile industry. Over 107 kg of dye are consumed annually by the global textile industry, with 90% of that amount reportedly ending up on textiles. Accordingly, the global textile sector releases 1,000 tons or more of dyes into waste streams annually (Robinson et al., 2001). Though primarily raw materials for textile industry are natural elements, various chemicals, pigments are added later in the various processes to reach at desired end product. Industry uses in large amount of water for the process

and output wastewater which is high in colour, Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), pH, turbidity, temperature, toxics etc are directly discharged in nearby water sources causing troubles to nature. When dye-containing wastewater is released into natural streams and rivers, it seriously harms aquatic life, the food chain, and the environment's aesthetic qualities. Because dyes absorb and reflect sunlight, they can hinder bacterial growth and aquatic plants' ability to perform photosynthesis. The issues worsen since the dyes' intricate aromatic structures make them inefficient against heat, light, bacteria, and even oxidizing agents, and their degradation becomes crucial (Pearce et al., 2003). Depending on the exposure contact time and dye concentration percentage, dyes can have detrimental and/or persistent impacts on exposed species. Allergy, skin irritation, cancer, mutation, and other conditions can be brought on by dyes. These are therefore a major problem since they endanger both human health and the purity of the water. One of the most important treatments for waste water is the elimination of dyes since their direct sight frightens the nearby and downstream living things. Nevertheless, removing color from textile effluent using less expensive and more eco-friendly technology remains a significant challenge. Industrial wastewater can be treated chemically, biologically, or physically to remove color (Elsagha A. et al., 2013).The process of waste-water treatment can be selected by some requirements such as cost, characteristics and availability of resources. Physicochemical techniques are widely used primary treatment of wastewater in various industries. These techniques include adsorption, chemical reaction, filtration, ion-exchange, coagulation/flocculation reverse osmosis, electro dialysis etc (Velmurugan et al., 2011). Adsorption is quickly becoming one of the most popular treatment strategies for aqueous wastewater. According to Kapdan and Kargi (2002), the adsorption process has several benefits, including the potential for low-cost regeneration, the availability of well-known process equipment, sludge-free operation, and sorbate recovery. Because of its high adsorption capacity, surface reactivity, micropore structures, and vast surface area, activated carbon is frequently utilized as an adsorbent for dye removal.The sustainability of this technique depends on the availability of cost-effective, high-quality adsorbents. While activated carbon has great potential for treatment and adsorption purposes, commercially available options are often pricey due to the use of non-renewable and expensive materials like coal, which is not ideal for pollution control. As a result, there has been a growing interest in producing activated carbon from renewable and more affordable sources, primarily industrial and natural by products, especially for wastewater treatment applications. However, this conversion process is necessary to turn natural materials into activated carbon. Natural waste materials, which are typically of low economic value and can be challenging to dispose of, play a significant role in this context (Geopaul, 1980).Researchers have found a variety of inexpensive sorbents for wastewater dye removal.These include the following Babassu coconut mesocarp (Vieira et al., 2009), coconut husk sorbents (Jain and Shrivastava, 2008; Low and Lee, 1990; Gupta et al., 2010), coconut shell fiber, (de Sousa et al., 2010; Babel and Kurniawan, 2004), coconut coir pith (Namasivayam et al., 2001), coconut bunch waste (Hameed et al., 2008), palm kernel fiber (El-Sayed, 2011), rice husk residues (Gupta et al., 2006; Lakshmi et al., 2009), sawdust (Batzias and Sidiras, 2007; Khattri and Singh, 2009), peanut shell (Tanyildizi, 2011), orange peels (Khaled et al., 2008; Arami et al., 2006), and waste tea leaves. The current research examines how effective unactivated lemon seed powder and moringa seeds are when used as low-cost adsorbents for the extraction of methylene blue dye. Methylene blue (MB) is the most used dye for coloring silk, wood, and cotton. It can potentially inflict eye burns that are dangerously damaging to the eye of both humans and animals. Inhaling it can trigger episodes of tachypnea or dyspnea, while orally consuming it can give a burning sensation and possibly lead to methemoglobinemia, in addition to nausea, vomiting, and sweating. Moringa seeds are gathered from overripe drums or moringa, while lemon seeds are collected from the neglected lemons at the citrus juice facility because not only are they deemed to be waste, they are also byproducts.

MATERIAL AND METHOD

Materials

Lemon and Moringa seeds were selected as adsorbents. Because they are readily available locally and are free of charge. We bought methylene blue (MB) from a chemist. With a molecular weight of 319.85 g/mol, MB's formula is C14H18N3SCI. It was not further purified before use. PH was adjusted using H2SO4 and NaOH. Every reagent was of analytical quality. The experiment was conducted using water that had been double distilled. The FTIR spectrophotometer (3000 Hyperion microscope with vertex with 80 FTIR system), UV–visible spectrophotometer

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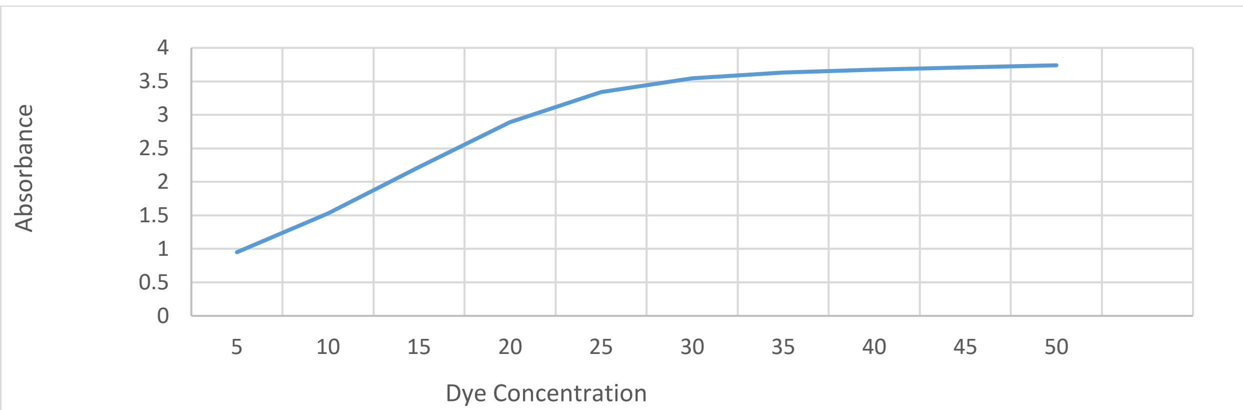
(SpectroquantPharo 300), mechanical shaker 20–880, weighing balance, pH meter-16, and sieves of around 300–600 μm size are among the tools used for the job.

Preparation of adsorbent

Moringa seeds were gathered from fields, and lemon seeds were gathered from a juice center. They were cleansed with distilled water after choosing the adsorbent, and the clean water was then used as wash water. Following a week or so of sun drying, it was ground into a powder using a home grinder. To get particles in this range, it was sieved via a 450–600 μm sieve. Experiments were conducted using particles retained on a 600μm sieve. Before being used for adsorption experiments, this produced powder was kept in a plastic container. Adsorption studies were conducted without any physical or chemical manipulations.

Preparation of adsorbate

In a one-litre volumetric flask, 1.0 g of dye was dissolved in double-distilled water to create the dye stock solution, which was then made to a concentration of 1 g L-1. The calibration curve below was created using the working solutions, which were made by precisely diluting the dye stock solution to the required starting concentrations (5–50 mg L-1).



Graph 1: Calibration curve of methylene blue on spectrophotometer.

Batch Adsorption experiments

The goal of the batch adsorption tests was to determine the best way to remove colour from textile waste water. Initial values of 10, 20, and 30 mg/l were tested. By doing the same experiment under various conditions, the equilibrium time and ideal adsorbent dosage were ascertained. For all three concentrations, the effects of contact time, dye concentration, adsorbent quantity, and pH of methylene blue (MB) adsorption were examined. The 250 ml flask used for the batch adsorption studies had a total solution capacity of 50 ml throughout the duration of the experiment. A mechanical shaker was used to shake the flasks at 150 rpm. At pH 6 and an adsorbent dose of 0.5 g, the impact of time on the elimination of methylene blue dye was investigated for intervals of 15, 30, 45, 60, 75, and 90 minutes. After that, samples were taken out for examination using a UV visible spectrophotometer, which tracked changes in absorbance at room temperature at λmax 651 nm. By adjusting the adsorbent dose range from 0.1 g to 0.8 g in 50 ml solution, the impact of adsorbent dose on the elimination of methylene blue dye was examined. Shaking the dye solution until the pH 6 equilibrium time is reached yields it. pH values ranging from 3.0 to 11.0 in a 50 ml dye solution for equilibrium time were used to examine the impact of pH on the elimination of methylene blue dye. To alter the pH values, 0.1 M HCl and 0.1 M H_2SO_4 were used. At varying dye concentrations, the impact of dye concentration on methylene blue dye removal was investigated. For the investigation, dye concentrations ranging from 5 mg/l to 50 mg/l were used. Following the adsorption experiment, centrifugation was used for 10 to 15 minutes at 3000 rpm to separate the adsorbent and the supernatant. Samples were then taken out for examination using a clinical syringe, and using a UV visible spectrophotometer, absorbance variations at λmax 651 nm were monitored at room temperature to determine the amount of dye left. The following is the amount of dye absorbed per gram of adsorbent (qe)

$q_e = \frac{V}{M}(C_e - C_0)$……………………………(1)

The percentage removal (R) was calculated using Eq. (2)

$\%R = \frac{Ce - C0}{C0} X100$……………………… (2)

Where, Co and Ce are the initial and equilibrium MB concentrations respectively (mg L^{-1}), V is the MB solution volume (L) and m is the mass of the adsorbent (g). The equilibrium data were evaluated using the Langmuir and Freundlich isotherm and characteristic parameters for both isotherms were determined.

Spectral analysis

An FTIR spectrophotometer (3000 Hyperion Microscope with Vertex 80 FTIR System) was used to record the spectra of lemon seed powder and Moringa seed powder in the 4500–500 cm-1 range. The IR graph's bands and stretch vibrations indicate that both samples include a variety of functional groups, including hydroxyl, aromatic, aliphatic, and carboxylic acid groups. Every peak exhibits a shift to a new wavelength, indicating that the functional groups are involved in the methylene blue adsorption process.

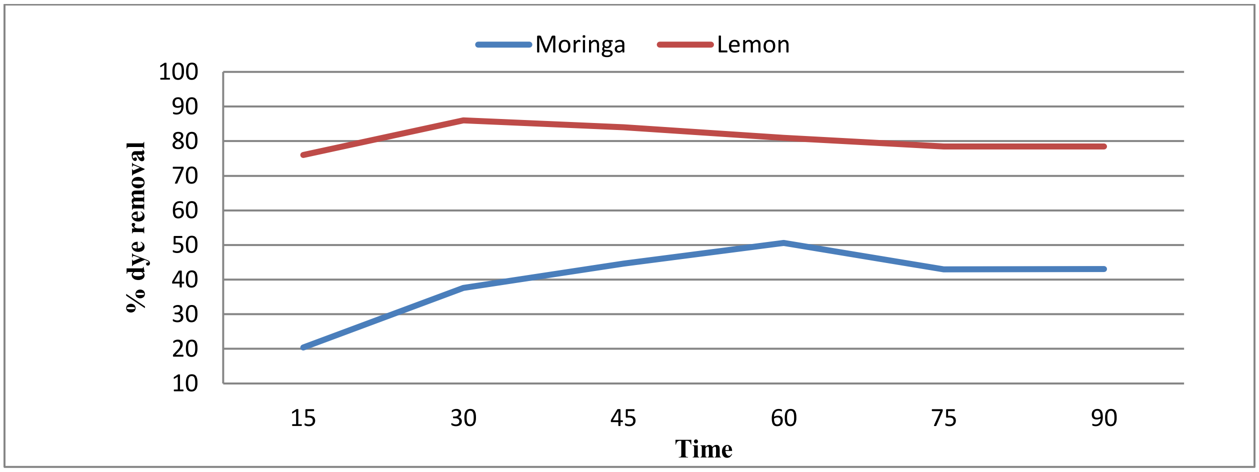
RESULTS AND DISCUSSION

The effectiveness of adsorbents on the removal capacity of methylene blue dye was compared. Batch experimentation served as the foundation for this. Adsorption was examined in relation to starting dye concentration, duration, pH, and adsorbent quantity. At a starting concentration of 20 mg/l, the adsorbent's ability to remove color is compared.

Effect of time

Experiments were conducted at three distinct concentrations (10 mg/l, 20 mg/l, and 30 mg/l) at pH 6 in order to examine the impact of time on the removal of methylene blue dye. It is evident from the comparing results that time is a significant factor in the color removal adsorption process.

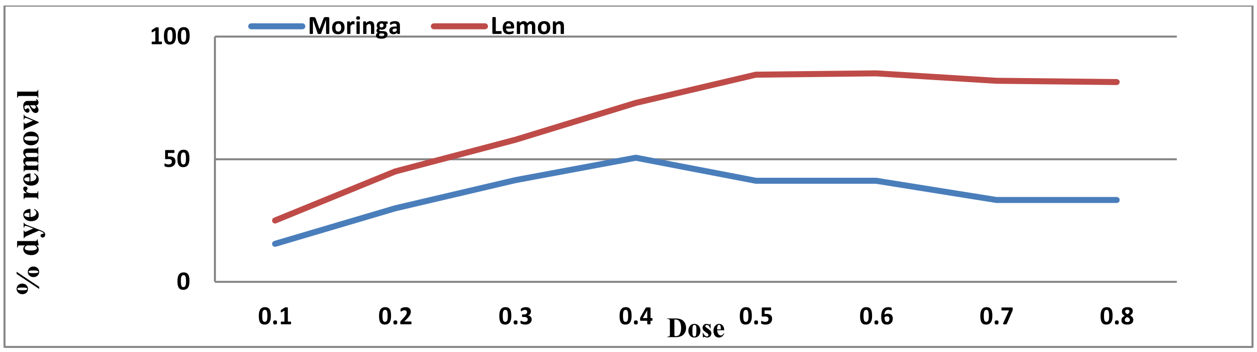
It was shown that dye uptake was rapid during the first few seconds of contact time, then it approached equilibrium and steadily increased for all three concentrations. The Moringa seed powder's color removal effectiveness peaked at 60, 60, and 75 minutes for dye concentrations of 10 mg/l, 20 mg/l, and 30 mg/l, respectively. At 60 minutes, it removes dye with a maximum efficacy of 75% for 10 mg/l. The minimal efficiency is 50.6% at a concentration of 20 mg/l and 38.34% at a concentration of 30 mg/l. The effectiveness of the lemon seed powder in removing color was demonstrated at 45 minutes for a dye concentration of 10 mg/l and at 30 minutes for both 20 mg/l and 30 mg/l. At 45 minutes, it removes dye with a maximum effectiveness of 94.5% for 10 mg/l, 86% for 20 mg/l, and 82.33% for 30 mg/l concentration. It is evident from the results that lemon seeds are far more effective than powdered Moringa seeds. Moringa seed has an efficiency of 50.6%, while lemon seed powder has an effectiveness of 86%. While Moringa seeds reach their maximum efficiency at 60 minutes, lemon seeds exhibit increased efficiency very early on, at 30 minutes.



Graph 2: Comparative results for efficiency of both adsorbents with respect to time

Effect of Adsorbent dose

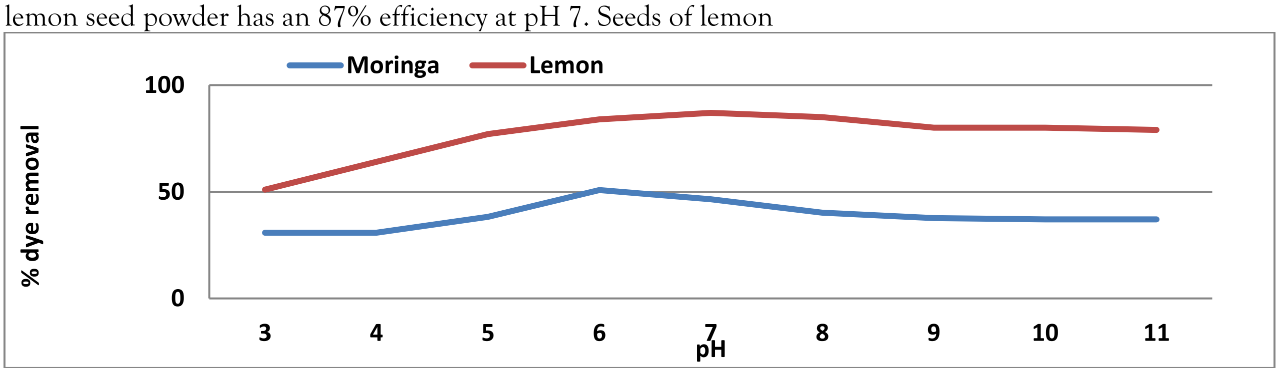
Because it establishes the adsorbent's capacity for a specific initial dye solution concentration, the adsorbent dosage is a crucial element in adsorption investigations. For all three concentrations, it is discovered that additional dosage increases cause the adsorption of dye to diminish or almost achieve equilibrium. Due to adsorption sites overlapping or aggregating, which lengthens the diffusion channel and reduces the overall adsorption surface area accessible to the dye. While Moringa seed yields an efficiency of 50.6% at 0.4 g in 60 minutes, lemon seed was shown to be highly effective with a color removal effectiveness of 84.5% at an adsorbent dose of 0.5 g in 30 minutes. Moringa seed powder only yields 50.6% efficiency, even though it reaches its peak at a lower adsorbent dose (0.4 g) than lemon seed powder. Up to a certain point, the adsorbent's capacity to remove dye grows as the dose does. We can infer from the graph below that adsorption slows down and approaches equilibrium after the ideal adsorbent dose is reached. For both adsorbent powders, it was discovered that adsorption was decreasing as the dose increased.



Graph 3: Comparative results for efficiency of both adsorbents with respect to adsorbent dose.

Effect of pH

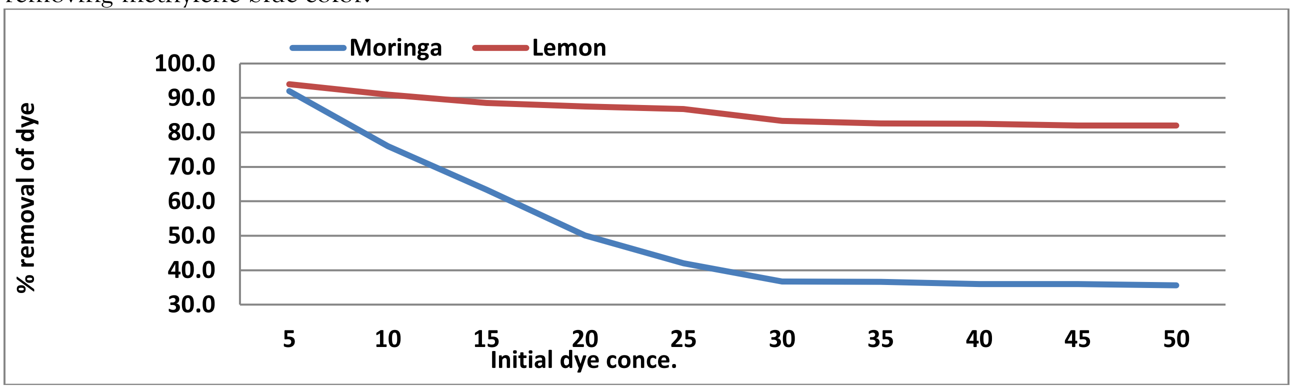
In adsorption research, particularly dye adsorption, the pH factor is extremely important. Both the ionized dye molecules and the charges on the adsorbent's surface transfer electrostatic charges, and the pH medium regulates the strength of these charges. Consequently, an aqueous medium's pH will affect the rate of adsorption. Color removal was examined at pH values ranging from 3 to 11 in order to see how pH affected the powdered Moringa seed's ability to remove color. To alter the pH values, 0.1M HCl and 0.1M H2SO4 were used. The presence of extra H+ ions competing with dye cations may be the cause of lower MB adsorption at very acidic pH values. The number of negatively charged surface sites on the adsorbent increased as the pH of the solution rose, which could lead to an increase in the adsorption of cationic dye molecules because of electrostatic attraction. Within the pH range of 3 to 11, the percentage of adsorption changed, suggesting that the chemical reaction between the adsorbent and dye molecules also influenced dye adsorption in the solution in addition to the electrostatic process. While color removal is more effective in near-alkaline conditions, it is less effective in acidic conditions. It is evident from the results that both adsorbents function best close to or at neutral pH. Moringa seed powder has a 50.8% effectiveness at pH 6 while lemon seed powder has an 87% efficiency at pH 7. Seeds of lemon



Graph 4: Comparative results for efficiency of both adsorbents with respect to pH

Effect of initial dye concentration

The original dye concentration has a significant impact on the percentage of dye removal. The direct relationship between the dye concentration and the accessible binding sites on an adsorbent surface determines the impact of the initial dye concentration factor. In general, when the initial dye concentration rises, the percentage of dye removal falls. This could be because the adsorption capacity increases as the initial dye concentration rises, or it could be because the adsorption sites on the adsorbent surface get saturated. Greater initial dye concentrations will result in a greater residual concentration of dye molecules. Lower concentrations result in a low ratio of starting dye molecules to available adsorption sites, which causes fractional adsorption to become independent of starting concentration .As is well known, the initial dye concentration affects how well dye is removed. The graph below illustrates how the percentage of dye removal falls as the starting dye concentration rises. Both adsorbents' removal effectiveness declines with increasing starting concentration. At 5 mg/l, the highest efficiency of 94% for lemon seed and 92% for Moringa is noted. At a dye concentration of 50 mg/l, the minimum efficiency for lemon seed is 82%, whereas for Moringa seed, it is 35.6%. Moringa seed powder's effectiveness as an adsorbent was significantly diminished. Based on a comparison of the aforementioned findings, lemon seed powder outperforms Moringa seed powder in terms of removing methylene blue color.



Graph 5: Comparative results for efficiency of both adsorbents with respect to initial dye concentration

Adsorptoin isotherm

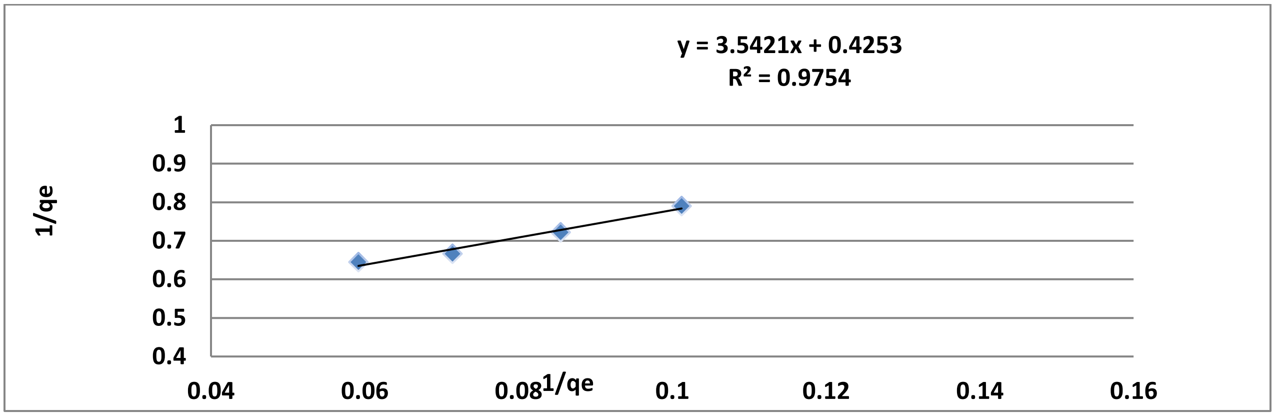
Important models for describing adsorption behavior are provided by adsorption isotherms. It provides an understanding of the nature and mechanism of the adsorption process as well as the interactions between the adsorbent and the adsorbate. To determine the adsorption capacity of different adsorbents, it is important to examine isotherm data. Two equilibrium isotherms were examined in order to study the adsorption isotherm: Freundlich and Langmuir isotherms are used to fit experimental data in adsorption studies in order to determine the degree and amount of favorability of adsorption. The Langmuir model was created under the presumption that the adsorbate species would form a monolayer on the adsorbent's surface. In order to assess the adsorbent's adsorption effectiveness, the Langmuir isotherm must be studied. Enhancing the operating conditions for efficient adsorption is another benefit of this study. The graph below displays the plots of 1/Qe against 1/Ce for both adsorbents. Plotting of the experimental data resulted in an equation that was compared to the y = mx + c equation. R2, KL, and q max are calculated using the graph values and are shown in Table No. 1. The system fits the isotherm, as shown by the graph's formation of a straight line. Below is the Langmuir equation.

$$q_e = \frac{q_{max} \; Kl \; Ce}{1 + Kl \; Ce} \quad \text{.............................}(3)$$

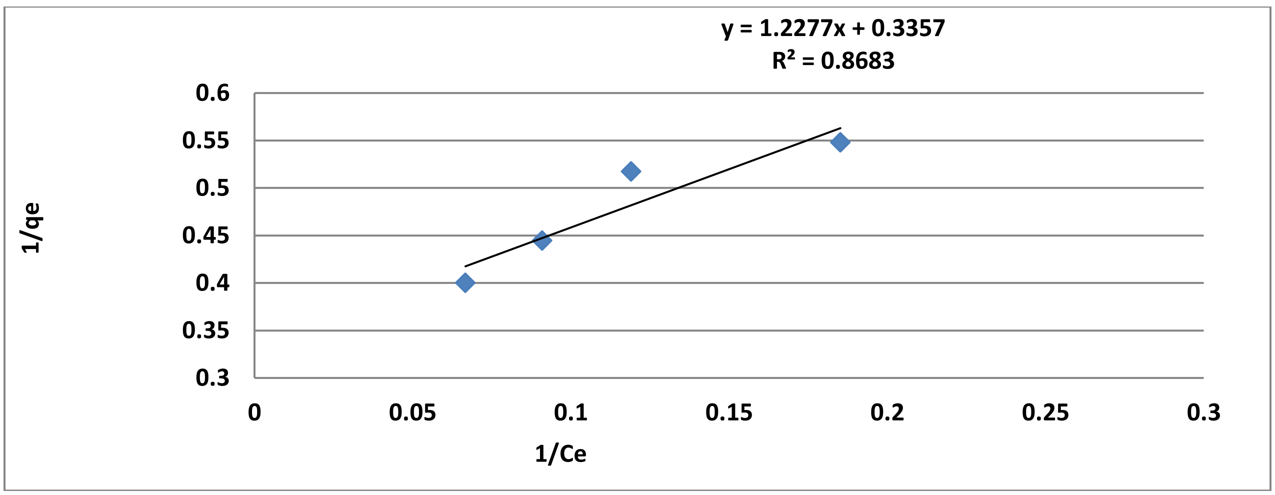
This can be linearized to

$$\frac{1}{qe} = \frac{1}{q_{max} \; Kl \; Ce} + \frac{1}{q_{max}} \quad \text{.............................}(4)$$

Where, Ce is the equilibrium concentration (mg/L), qe is the amount of dye adsorbed per unit mass of adsorbent at equilibrium (mg/g), q_{max} is the theoretical maximum adsorption capacity (mg g^{-1}), K_L is the Langmuir isotherm constant (L mg^{-1}).



Graph 6: Graphical Representation of Langmuir Isotherm for Moringa seed powder.



Graph 7: Graphical Representation of Langmuir Isotherm for lemon seed powder.

Table 1: Langmuir Constants for various adsorbents

Adsorbent	Langmiur Constant		
	Q_{max}	KL	R2
Moringa seed	2.351	0.12	0.9754
Lemon seed	2.9788	0.273	0.868

When designing adsorption systems, the equilibrium adsorption isotherms are crucial. The Freundlich isotherm may provide adequate equilibrium adsorption data. The equation for Freundlich is provided as

$q_e = K_f Ce^{1/n}$……………………………(5)

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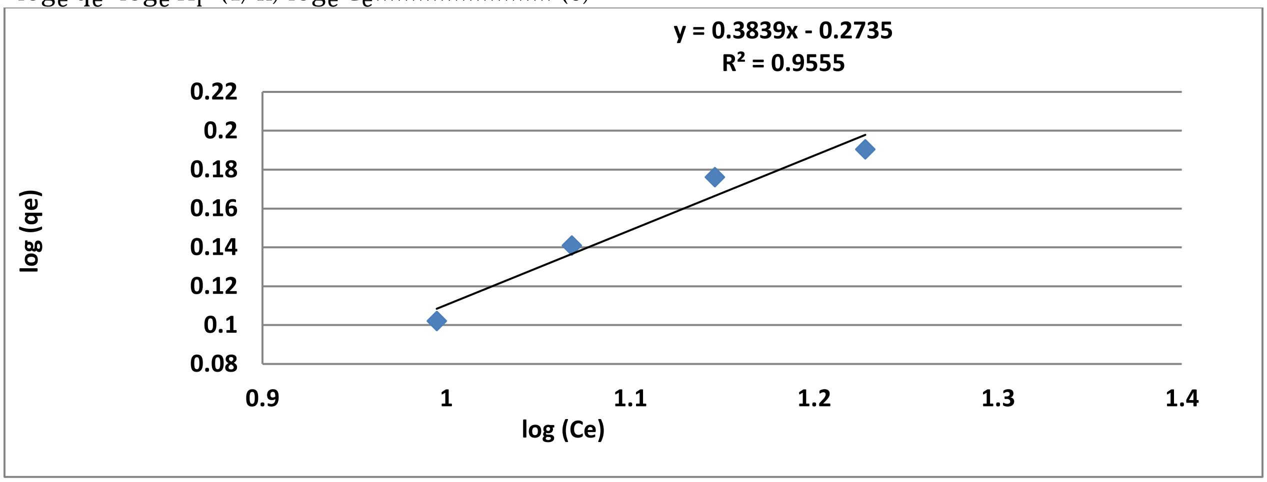
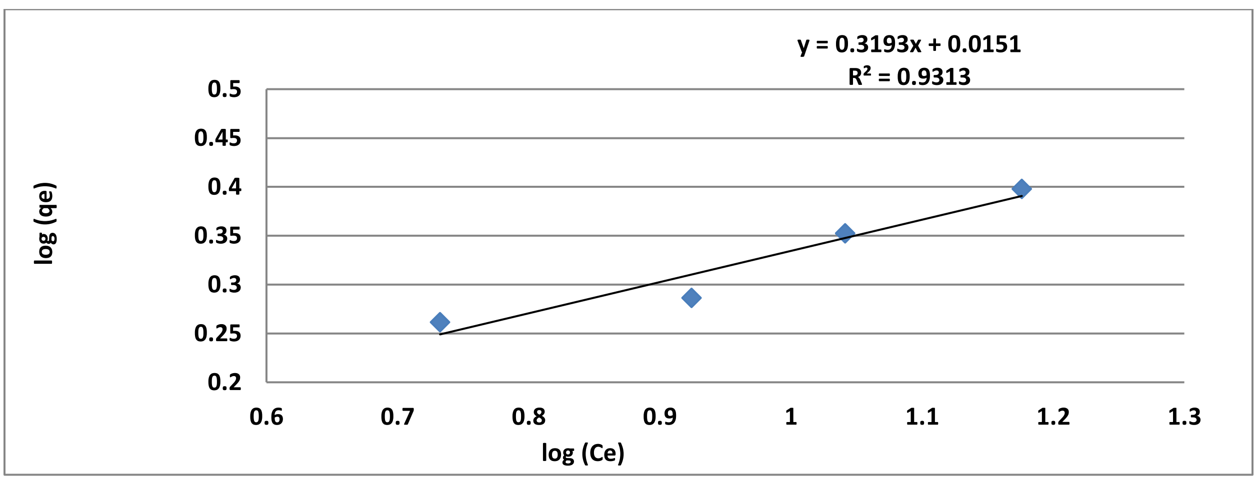
where Ce is the equilibrium concentration (mg L^{-1}), Kf is the Freundlich adsorption constant associated with the adsorbent's adsorption capacity, n is a dimensionless constant that can be used to explain the extent of adsorption and the adsorption intensity between the solute concentration and adsorbent, and qe is the amount of dye adsorbed per unit mass of adsorbent at equilibrium (mg/g). The Freundlich equation's linear form is typically written as

$\log_e q_e = \log_e K_f + (1/n) \log_e C_e$........................ (6)



Graph 8: Graphical Representation of Freundlich Isotherm for Moringa seed powder



Graph 9: Graphical Representation of Freundlich Isotherm for lemon seed powder.

Graphs 8 and 9 display the plots of Log Qe vs Log Ce, and the equation derived from the graph is contrasted with the y = mx + c equation. The system is said to fit the Freundlich Isotherm when a straight line forms. The data shown in table No. 2 were generated by applying the graph's value to the isotherm equation; the kind of the isotherm is indicated by the value of R. For both adsorbents, the value of R was deemed favorable. This once more demonstrated that, in the circumstances employed in this investigation, the Langmuir and Freundlich isotherms were advantageous for the adsorption of MB on natural adsorbent.

Table 2: Freundlich Constants for various adsorbents

Adsorbent	Freundlich Constant		
	1/n	K_f	R^2
Moringa seed	0.3839	0.53272	0.9555
Lemon seed	0.3193	1.0353	0.9313

The value of R indicates the type of the isotherm. If $R > 1$ isotherm is unfavorable, If $R = 1$ isotherm is Linear, if $0 < R < 1$ isotherm is Favorable or if $R < 0$ isotherm is Irreversible.

CONCLUSION

According to the current study, methylene blue dye could be eliminated from aqueous solutions using powdered moringa and lemon seeds. It was discovered that methylene blue adhered firmly to the surfaces of both adsorbents. Fourier transform infrared (FTIR) spectroscopy was used to examine the functional groups found in the proximate composition of the lemon and moringa seeds. According to the results of this investigation, lemon seed powder is a more potent natural adsorbent than moringa seed powder at removing MB color. At room temperature, lemon seeds remove MB dye with an efficiency of 86%, which is significantly higher than that of moringa seeds, which only remove roughly 50%. The optimal pH for moringa seed powder is 6, whereas the optimal pH for lemon seed is 7. For both adsorbents, dye removal efficiency either stabilizes or marginally declines as pH rises above 7. Lemon seed processed under nominal treatment was determined to be more effective than moringa seed, according to the isotherm study. The good linear correlation coefficient values show that adsorption parameters derived from Langmuir-Feyerlich isotherms are helpful for explaining the mechanisms of the adsorption process.

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SUMMARY

this research explored the use of Moringa seed powder and lemon seed powder as low-cost, natural adsorbents for removing methylene blue dye from synthetic textile wastewater. Variables studied included contact time, dye concentration, pH, and adsorbent dose.

ABBREVIATIONS

MB – Methylene Blue.
BOD – Biological Oxygen Demand.
COD – Chemical Oxygen Demand.
pH – Potential of Hydrogen.
FTIR – Fourier Transform Infrared (Spectroscopy).
UV – Ultraviolet.
rpm – Revolutions Per Minute.
qe – Amount of dye adsorbed per unit mass of adsorbent at equilibrium (mg/g).

Ce – Equilibrium dye concentration (mg/L).
Co – Initial dye concentration (mg/L).
V – Volume of dye solution (L).
m – Mass of adsorbent (g).
qmax – Maximum adsorption capacity (mg/g).
KL – Langmuir adsorption constant (L/mg).
Kf – Freundlich adsorption constant.
1/n – Freundlich adsorption intensity parameter.
R^2 – Coefficient of determination.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

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