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Removal of Malachite Green from Aqueous Solution using Chemically Treated Activated Carbon

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Abstract

The increasing discharge of dye waste water from Pakistan textile industries causes serious environmental problems. This study focus to explore an efficient method to remove malachite green dye through chemically treated activate carbon derived with (coal waste and calcium silicate). This study examines the effectiveness of activated carbon under controlled conditions and looks into how acid and alkali modifications affect the material adsorption capabilities. Several Batch adsorption experiments have been conducted by varying factors like adsorbent type and contact time. Langmuir and Freundlich model isotherms have been used to access the adsorbents performance, and pseudo first order and second order have been used to investigate adsorption. The acid treated activated carbon gives the highest dye removal efficiency attaining 80% removal efficiency in a short period of time. While the kinetic data more closely matched the pseudo-second-order model, indicating that chemisorption is the predominant process, the equilibrium data better fit the Langmuir isotherm, indicating monolayer adsorption onto a homogenous surface. Because of its increased surface area and pore structure, these data demonstrate how well acid-treated activated carbon adsorbs dye molecules. This study helps to develop cost-effective and environmentally friendly wastewater treatment strategies in dye-intensive industries. In conclusion, acid-treated activated carbon offers a promising, sustainable solution for industrial dye wastewater treatment. Its high removal efficiency and rapid adsorption kinetics make it a suitable candidate for large-scale applications.

Keywords: Adsorption, Activated Carbon, Malachite Green, Textile Industry Wastewater

1. Introduction:

Although having access to safe and clean water is a fundamental human right, millions of people worldwide still suffer from waterborne diseases due to inadequate sanitation and contamination [1]. According to literature, unsanitary water is responsible for approximately 1.8 million deaths annually, highlighting the urgent need for effective water safety management [2]. While water suppliers in over 20 countries have adopted World Health Organisation (WHO) guidelines to improve water sanitation, many nations, including Pakistan, still face significant challenges in addressing water pollution and ensuring safe water for their populations. Gleick 2014 [3]. One of the most pressing concerns in water contamination is the existence of dyes, which are extensively utilised in sectors like paper and textiles, leather and laboratories [4]. These dyes not only affect the aesthetic quality of water but also pose serious environmental and health risks [5]. Even at concentrations as low as 1 ppm, dyes can make water visibly coloured, leading to public perception of contamination and rendering it unfit for consumption [6]. More than 0.7 million tons of coloured products are produced each year using more than 10,000 commercially available dyes, resulting in high biological oxygen demand (BOD) and persistent dye residues in

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water bodies [7]. This contamination has determined effects on aquatic ecosystems and human health, making it imperative to develop effective methods for dye removal from wastewater [8].

Because of its deep shade and affordability, Malachite green (MG), a synthetic triphenylmethane dye, is frequently used in the textile and dyeing industries to colour leather, jute, and silk [9]. However, Malachite green is a significant environmental pollutant due to its toxicity, mutagenicity, and poor biodegradability [10]. It builds up in aquatic life and can be harmful to human health, including genotoxicity and respiratory consequences [11], [12]. Eliminating Malachite green from wastewater is essential due to its chemical stability and resistance to biological treatment. The effectiveness of activated carbon in removing Malachite green from textile effluents is examined in this work.

In Pakistan the textile industry is a major contributor to water pollution, with composite units, bleaching and dyeing processes being the primary sources of wastewater discharge [13]. Despite the implementation of six key environmental protection laws by the Pakistani government, the textile sector continues to generate significant volumes of contaminated water, necessitating innovative and cost-effective solutions for wastewater treatment [8]. Chemical oxidation is one of the techniques that have been investigated for the removal of colours from wastewater, along with coagulation, membrane separation, electrochemical techniques and aerobic degradation [14]. However, these methods often face limitations such as high operational costs, generation of harmful by-products and inefficiency in treating complex wastewater [15]. Among these, adsorption has emerged as a highly effective and versatile technique for water purification [16]. Adsorption is not only cost-effective and easy to operate but also environmentally friendly, as it does not produce harmful by-products [17]. The most common adsorbent is activated carbon, which is particularly notable for its substantial surface area, Microporus structure and high adsorption capacity, making it ideal for removing dyes and toxic substances from water[18]. The use of coal-based activated carbon as an adsorbent to remove malachite green colour from wastewater from the textile industry is the main topic of this study[19]. Activated carbon's efficient kinetics, cost-effectiveness and ability to treat both acidic and alkali dyes make it a promising solution for wastewater treatment[20]. The study also explores the impact of chemical treatment (acid and alkali) on the adsorption efficiency of activated carbon[21], with acid-treated activated carbon demonstrating a superior performance in the removal of the dye [22]. By addressing the challenge of dye contamination in wastewater[23], this research aims to contribute to the development of sustainable and effective water treatment technologies, ultimately promoting environmental protection and public health [24].

2. Experimental Works:

2.1. Materials:

2.1.1. Malachite Green:

Malachite green (MG), also referred to as Basic Green 4, and is a synthetic triphenylmethane dye with a molar mass of 364.911 g /mol and the molecular formula $C_{23}H_{25}ClN_2$. It has strong cationic qualities and a high affinity for textile fibers due to its chemical structure, which consists of a core carbon bound to three aromatic rings that have been replaced with methylamino groups. The dye employed in this investigation had an average particle size of 50 microns and was purchased from Sigma Life Science (product number: 32754). In addition to being a biological stain and antifungal agent in fish farming, Malachite green finds extensive application in the textile, leather, paper, and aquaculture industries. However, because of its genotoxic, mutagenic, and carcinogenic qualities, it is regarded as dangerous. Research has connected chromosomal abnormalities to Malachite green exposure. There are major environmental and food safety concerns because its reduced form, leucomalachite green, has been found in exposed fish's muscles, liver, and kidney. With a pH range of 0.2 to 1.8, Malachite green may function chemically in extremely acidic environments.

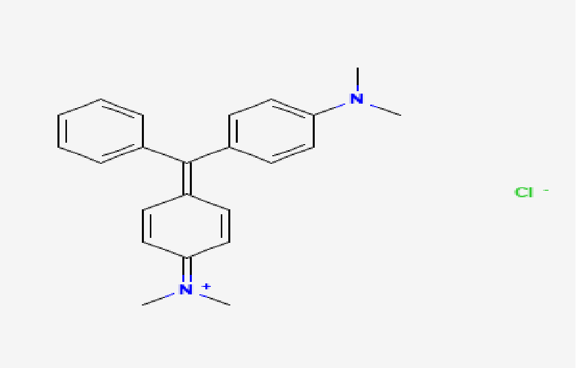

Figure 1: Chemical structure of Malachite green

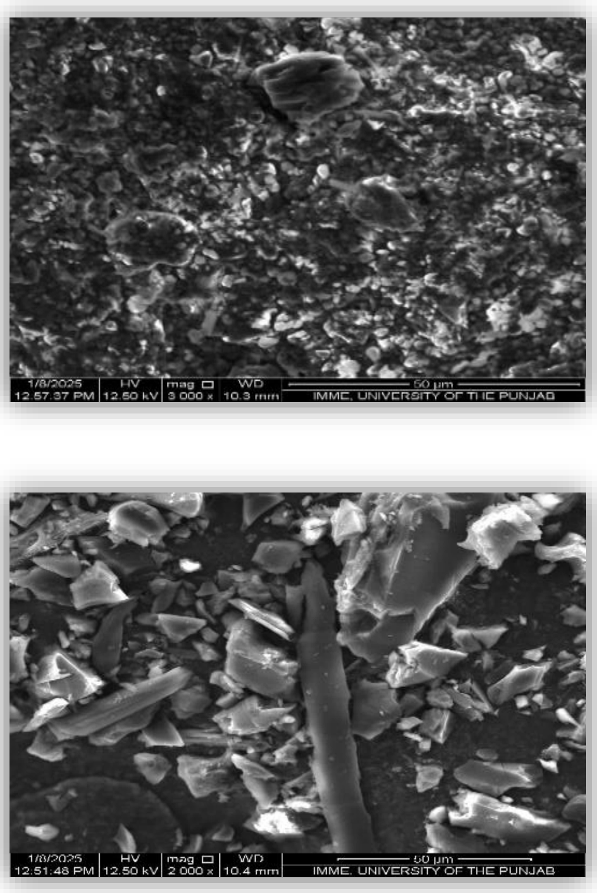

Figure 2: SEM image of Malachite green a) After adsorption b) Before adsorption

2.1.2. Activated carbon:

Al-Hayat & Company, located on Alamdar Road in Lahore, provided the Activated carbon, which has a surface area of more than 1000 m²/g. Commercial activated carbon derived from coal is widely used in wastewater treatment because of its high porosity, superior adsorption capacity, and capacity to remove organic pollutants, dyes, and heavy metals. Its recyclable nature improves environmental applications' sustainability even further.

2.2. Methodology:

2.2.1. Analysis of Malachite green in aqueous solution:

Using distilled water on a volume basis, the stock solution of Malachite green (MG) used in this study was made at a concentration of 1000 ppm (mg/L). Working standard solutions of 80, 60, 40, 20, and 10 ppm were made from this stock solution by serial dilution for use in experimental procedures. These standard solutions were examined using a UV-Visible spectrophotometer, and a calibration curve was created by plotting absorbance versus concentration (ppm) to guarantee precise quantification of MG in aqueous samples in accordance with standard analytical protocols.

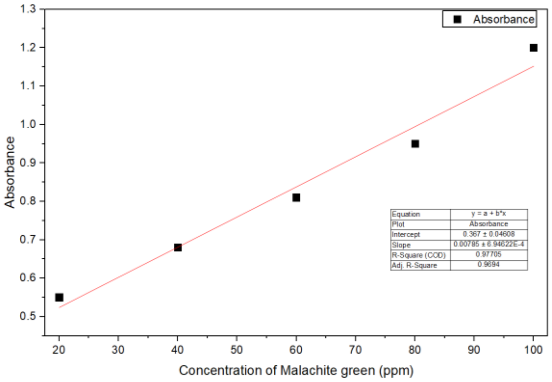

Figure 3: Calibration curve between Absorbance & Concentration (ppm)

3. Adsorption Experiments:

The study was conducted at the Punjab University's Institute of Chemical Engineering and Technology in Lahore. Based on volume, a stock solution of 1000 ppm malachite green solution was made. During Malachite green's adsorption onto activated carbon compounds, many adsorption properties were established, such as kinetic studies, adsorption isotherms, adsorbent dose, and pH. The kinetics of adsorption was characterized by establishing the adsorption isotherms and implementing several kinetic models, such as pseudo-first and pseudo-second order. To describe the adsorption isotherms and assess which model best fit this situation; adsorption models such as the Freundlich and Langmuir models were used.

3.1. Adsorption Kinetics:

The adsorption experiment was conducted using a one-liter flask containing a malachite green solution and a one-hundred-milliliter sample with a concentration of one hundred parts per million. A mass of 12.5 grams of adsorbent was introduced into the mixture. At five-minute intervals for the first half-hour and ten-minute intervals for the second half, samples were taken out. In order to guarantee the removal of small particles, the concentration of malachite green in each sample was assessed following filtration using Whatman filter paper and a zero-point-four-five micrometer nylon syringe filter (twenty-five millimeter diameter). It was possible to determine the adsorption equilibrium time by plotting the concentration at different time intervals against time.

Pseudo first order*:*

$$q_t = q_e * \left(1 - e^{\{-k_1 t\}}\right) \qquad \text{Eq (1)}$$

- q_t= Adsorption capacity at time t(mg/g)
- q_e= Adsorption capacity at equilibrium (mg/g)
- k = Rate constant of adsorption (1/min)
- t = Contact time (minutes)

Pseudo second order:

$$q_t = \frac{(q_e^2 * k_2 * t)}{(1 + q_e * k_2 * t)} \qquad \text{Eq (2)}$$

- q_t = Adsorption capacity at time t(mg/g)
- q_e = Adsorption capacity at equilibrium (mg/g)
- k_2 = Pseudo-second-order rate constant (g/mg·min)
- t = Contact time (minutes)

3.2. Adsorption Isotherms:

The duration of adsorption for every concentration of malachite green solution, which varied from 10 to 100 ppm, was calculated kinetically using a magnetic stirrer at a predetermined rpm. After extracting the samples and filtering them with Whatman filter paper and a syringe filter, the concentration at equilibrium C_e was resolute. The necessary isotherm for a certain model chemical was obtained by plotting q_e vs. C_e once the adsorption capacity was established. Eq (3) was used to calculate the adsorbent loading q_e (mgg-1) based on the samples' beginning and end concentrations**.**

- q_e = Amount of solute adsorbed at equilibrium per unit mass of adsorbent (mg/g)
- C_0= Initial concentration of solute in solution (mg/L)
- C_e = Equilibrium concentration of solute in solution (mg/L)
- V = Volume of the solution (L)
- m = Mass of the adsorbent (g)

3.2.1. Langmuir equation:

$$Qe = \frac{qmax\ KL\ Ce}{1+Kl*Ce} \qquad \text{Eq(3)}$$

Where:

- q_e = Amount of solute adsorbed per unit mass of adsorbent at equilibrium (mg/g)
- q_{max} = Maximum adsorption capacity corresponding to complete monolayer coverage (mg/g)
- K_l = Langmuir constant related to adsorption affinity (L/mg)
- C_e = Equilibrium concentration of solute in solution (mg/L)

Equation (4) represents the Langmuir isotherm, which assumes monolayer adsorption onto a surface with a finite number of identical sites. Here, q_{max} indicates the maximum adsorption capacity of the adsorbent, while K_l is equilibrium constant related to the affinity between the adsorbate and adsorbent. C_e is the concentration of the solute remaining in the solution at equilibrium, and q_e is the amount adsorbed per unit mass of adsorbent.

3.2.2. Freundlich equation:

$$qe = kCe^{\frac{1}{n}} \qquad \text{Eq(4)}$$

- q_e: Adsorption capacity at equilibrium (mg/g)
- C_e: Equilibrium concentration (mg/L)
- K: Freundlich constant (mg/g)(L/mg)^
- n: Heterogeneity constant (dimensionless)

3.3. Adsorbent Dosage's Impact on Malachite Green Loading onto Activated Carbon:

To investigate10 how adsorption capacity is affected by adsorbent dosage. In a 100 ml flask, solutions of malachite green at a concentration of 100 ppm were mixed with 2, 4, 6, 8, and 10 grams of alkali-treated activated carbon. The adsorption capacity of each dosage was assessed. For the malachite green solution to be adsorbed, only 2 grams of adsorbent are required, and increasing the dosage typically has little to no influence on the adsorption capacity.. It initially increases the adsorption capacity by filling the active sites; however, when the adsorbent's active sites decrease, its adsorption gradually decreases.

3.4. Effect of pH:

Following the establishment of the adsorbent dosage and equilibrium period, the pH was measured using a digital pH meter. To determine the pH, 0.05 normal solutions of HCl and NaOH were added drop by drop. The equilibrium capacities for alkaline, acidic, and neutral adsorption were ascertained .pH 6 gives the best adsorption capacity indicating increasing electrostatic attraction brought on by surface deprotonation. Base-treated activated carbon, on the other hand, showed less varied and lower adsorption, most likely as a result of preexisting surface negativity and restricted further charge generation at higher pH.

4. Results and Discussions:

4.1. Adsorption Kinetics:

Malachite green was used as a model contaminant to gauge adsorption kinetics. The equilibrium time was calculated in the study using concentrations of 100 ppm. Time-dependent estimations of the adsorption capacity were made. It was discovered that the adsorption process for alkali-treated activated carbon moved rather swiftly, achieving 70% to 80% of the equilibrium adsorption capacity in 40 to 50 minutes, while for acid-treated activated carbon, For simple activated carbon, 80% of the equilibrium adsorption capacity was reached in the first 40 minutes of the adsorption process, and the adsorption process was found to be fairly rapid, achieving 80% to 85% of the equilibrium adsorption capacity in 30 minutes[25], which proceeded more quickly than anticipated[26, 27]. To illustrate the variation in adsorption between beginning concentration and time, graph was created.

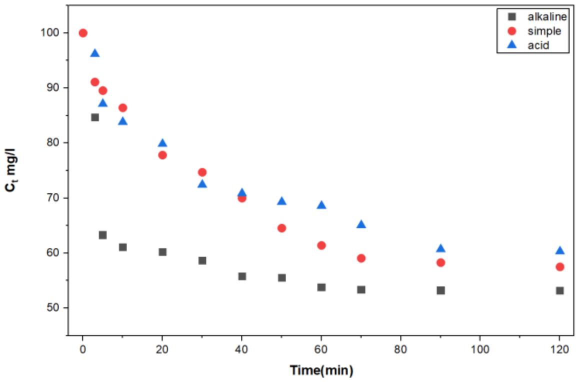

Figure 4: Adsorption kinetics for Simple, Acid, and Alkaline treated activated carbon on loading of Malachite green

The results show that oppositely charged molecules adsorb quickly due to electrostatic attraction and readily occupy active sites. In contrast, alkali base-activated carbon, having similar surface charges, experiences repulsion, resulting in slower adsorption. Simple activated carbon exhibits moderate performance with balanced adsorption time.

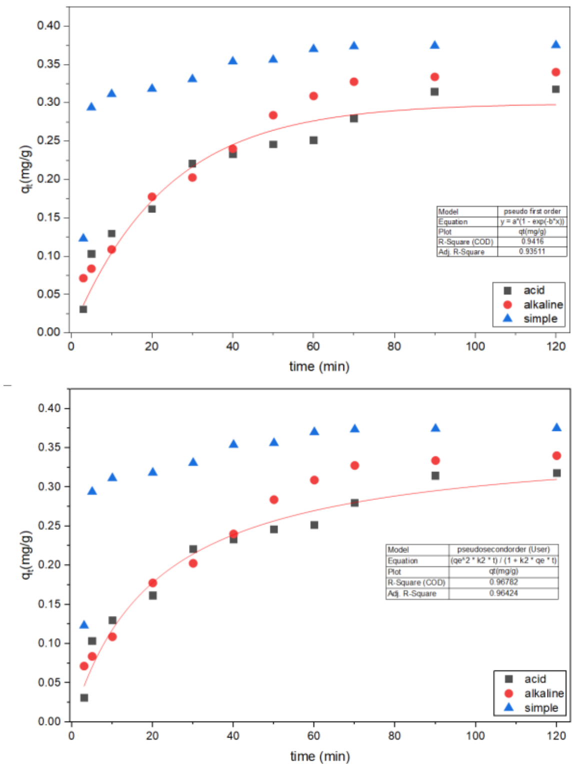

Figure 5: Adsorption kinetic data of Acid, simple and Alkali-treated Activated carbon upon loading Malachite green, a) pseudo first order & b) pseudo second order non-linear equation.

The kinetic rate constants and maximum rate adsorption capacity are evaluated through the models is shown in the Table1:

Table 1: Kinetic rate constant and maximum adsorption capacity for model compounds

Adsorbent	Pseudo 1st order model			Pseudo 2nd order model		
	K 1	Qmax(mgg-1)	R^2	K 2	Qmax (mgg-1)	R^2
Acid treated activated carbon	0.02468	0.3942	0.97	0.0489	0.4441	0.97
Simple activated carbon	0.11369	0.3599	0.86	0.3730	0.4009	0.87
Alkali treated activated carbon	0.02038	0.3251	0.95	0.0296	0.3851	0.96

The acid-treated activated carbon among the three adsorbents exhibited similarly high R^2 values (0.97) for both pseudo-first and pseudo-second order models, indicating that both models adequately capture the kinetics. Nevertheless, the pseudo-second order model exhibits somewhat larger R^2 for simple and alkali-treated activated carbon, suggesting that pseudo-second order kinetics which implies chemisorption better matches the data in general.

4.2. Adsorption Isotherm:

Using a magnetic stirrer set to a specific rpm, a kinetic study was conducted to ascertain the adsorption period for each Malachite green solution at different concentrations, ranging from 10 to 100 ppm. The samples were then extracted and filtered using a syringe filter and to ascertain the equilibrium concentration of C_e, use Whatman filter paper. A plot of q_e vs. C_e produced the necessary isotherm for a specific model molecule after the adsorption capacity was assessed. The findings indicate that the best adsorption capacity is provided by acid-treated activated carbon since the dye is basic in nature and the adsorbent's surface is charged with acid, which draws more and more malachite green particles to its active surface sites As a result of the force of repulsion, simple activated carbon has a modest rate of adsorption, whereas alkaline-treated activated carbon has a lower rate than any acidic and simple activated carbon.

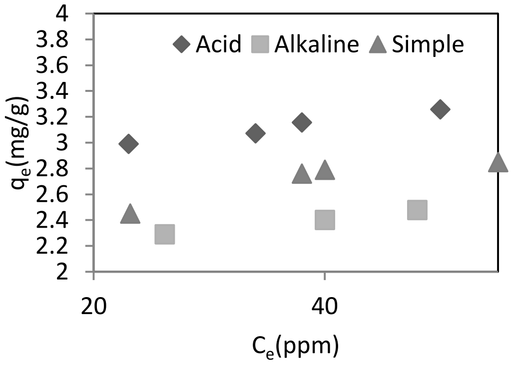

Figure 6: Comparison of adsorption isotherm models for the adsorption of Malachite green (100 ppm, 2 gram, 60 mins, 250C) using ordinary activated carbon and acid-treated activated carbon

4.2.1. Adsorption isotherm models (Freundlich & Langmuir):

Activated carbon treated with acid and alkali exhibited a superior fit (higher R²) to the Freundlich model, suggesting that chemical modification results in multilayer adsorption on heterogeneous surfaces. On the other hand, the Langmuir model fit untreated activated carbon better, indicating a more uniform surface with monolayer adsorption behaviour[28], as show in figure below,

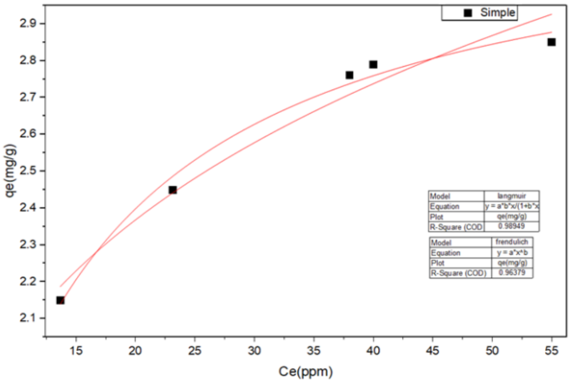

Figure 7: Adsorption isotherm models (Freundlich and Langmuir) for simple activated carbon applied to isotherm data for Malachite green

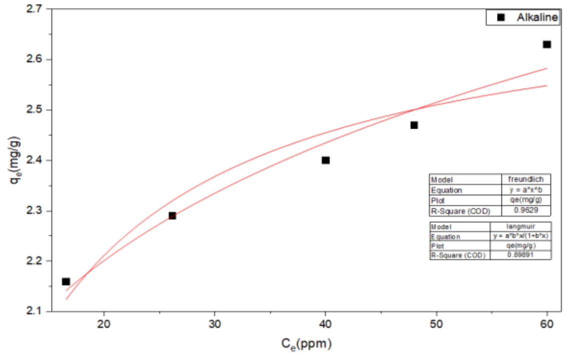

Figure 8: Adsorption isotherm models (Freundlich and Langmuir) for Alkaline-treated activated carbon applied to isotherm data for Malachite green

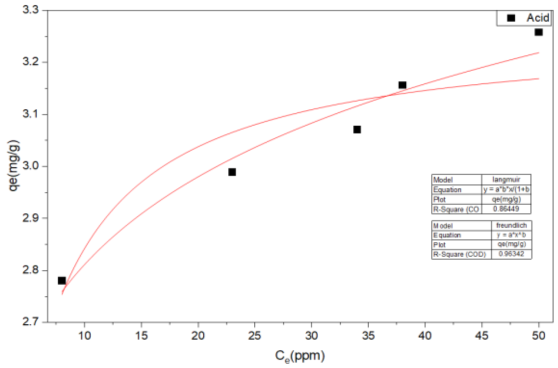

Figure 9: Adsorption isotherm models (Freundlich and Langmuir) for Acid treated activated carbon applied to isotherm data for Malachite green

Table 2: estimation of model parameters for adsorption of malachite green on chemically treated activated carbon

Isotherm model	Equation	Model parameters	Simple Activated carbon	Acid treated Activated carbon	Alkali treated Activated Carbon
Freundlich	$q_e = K_F C_e^{1/n}$	K_F	0.20	0.084	0.145
		n_F	1.26	2.3162	1.42433
		R^2	0.96	0.96	0.96
Langmuir	$q_e = \frac{K_L C_e}{1+bC_e}$	K_L	0.14	0.677	0.202
		b	3.12	3.262	2.75
		R^2	0.98	0.86	0.89

The use of the previously described models shows that the Freundlich model best describes the data for malachite green adsorption through activated carbon. In the following sequence, a number of models are useful for the adsorption of malachite green with activated carbon: More amiable than Langmuir

4.3. Effect of Adsorbent Dosage:

Different amounts (2, 4, 6, 8, and 10 grams) of chemically treated activated carbon, including simple, acid-treated, and alkali-treated varieties, were added to 100 ppm Malachite Green (MG) solutions in a 100 mL flask. According to the results of the experiment, the highest adsorption capacity for all three types of activated carbon was obtained with a two-gram dosage of adsorbent. Adsorption capacity was not appreciably increased by administering more adsorbent than two grams. The saturation of the solution's available dye molecules is the cause of the observed pattern. The number of active sites rises as the adsorbent dosage does, but the dye concentration stays constant. Thus, there is no appreciable improvement in adsorption capacity since the extra adsorption sites are left unused. Moreover, site overlap and particle aggregation may decrease the effective surface area available for dye adsorption at larger adsorbent dosages. Figure 9 below shows how adsorbent dosage affects adsorption capacity.

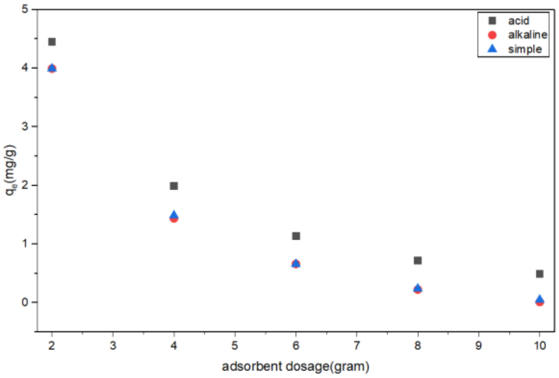

Figure 10: Effect of adsorbent dosage on the adsorption of Malachite green by acid-treated, simple, and alkali-treated activated carbon at pH 6 (100 ppm, 25°C).

4.4. Effect of pH on Malachite Green:

Following the establishment of the adsorbent dosage and equilibrium period, the pH was measured using a digital pH meter. To determine the pH, 0.05 normal solutions of HCL and NaOH were added drop by drop. The impact of pH on Malachite Green adsorption was examined for activated carbon that had been treated with both acid and base. The graph indicates that the adsorption capacity q_e of acidic Peaking at about (pH 6), activated carbon rose with pH, indicating increasing electrostatic attraction brought on by surface deprotonation. Base-treated activated carbon, on the other hand, showed less varied and lower adsorption, most likely as a result of preexisting surface negativity and restricted further charge generation at higher pH. These findings demonstrate that the pH-responsive adsorption behaviour of activated carbon is considerably enhanced by acid treatment [29, 30].

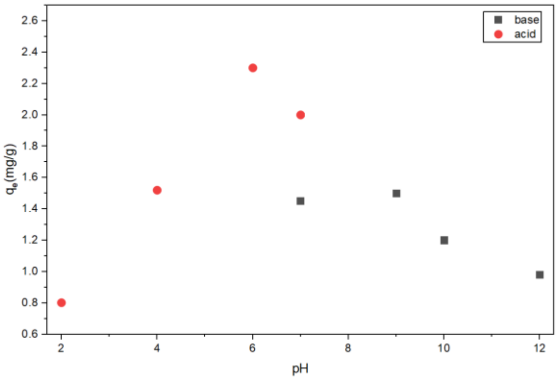

Figure 11: Acid and Alkali-treated Activated carbon's adsorption capacity for Malachite green at varying pH values: an impact of pH (100ppm, 2 gram, 25^{0}C, 0.05MHCl, 0.05M NaOH)

5. Conclusion:

Activated carbon is a useful substance for eliminating contaminants from aqueous systems because of its high adsorption capability and huge surface area. Although its efficacy in eliminating Malachite Green dye has been the subject of numerous investigations, there are still few comparative analyses of chemically modified activated carbon under ideal circumstances. By contrasting the adsorption capacities of activated carbon that has been acid-treated, alkali-treated, and untreated at pH 6, which has been determined to be the ideal pH for maximal dye removal, this work fills that gap. Superior adsorption kinetics was demonstrated by acid-treated activated carbon, which removed 85% of Malachite Green in 25–30 minutes and reached equilibrium in less than 40 minutes. Stronger dye binding was made possible by the addition of functional groups and active sites through acid modification, which is responsible for this improved performance. The Freundlich model, which suggests multilayer adsorption on heterogeneous surfaces, was found to best suit the acid- and alkali-treated samples by isotherm modeling. On the other hand, untreated activated carbon fit the Langmuir model better, which suggests monolayer adsorption on a more uniform surface. Because of the electrostatic repulsion between the basic dye and the negatively charged carbon surface, alkali-treated activated carbon showed slower adsorption kinetics and a reduced capacity. The adsorption performance of untreated activated carbon was modest, although it took longer to reach equilibrium. The deprotonation of surface functional groups, which improved electrostatic interactions with the cationic dye, is probably what caused the increased removal efficiency at pH 6. Furthermore, dosage studies demonstrated that 100 mL of a 100 ppm Malachite Green solution could be efficiently treated with 2 grams of acid-treated activated carbon. All things considered, acid modification greatly enhances activated carbon's adsorption capabilities, providing a quick, effective, and economical method of dye removal in wastewater treatment. Future studies should concentrate on the regeneration and reusing of modified activated carbon, assess how well it works in hybrid or continuous flow systems, and test its effectiveness in actual wastewater situations.

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