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Comparison between the results of the Langmuir and Freundlich isotherm for Removal of Methylene blue dye from Colored Wastewater

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Abstract

The capacity of eco-friendly biosorbents low cost a biosorbent for methylene blue dye (MBD) removal from wastewater is investigated in this work. In order to assess the potential treatment of industrial aqueous solutions by Activated Carbon, Palm leaf and Pomegranate peel, a series of laboratory batch flow experiments were conducted . Batch studies were carried out to investigate the effects of pH dye solution, contact time, biosorbent dosage, beginning dye concentration, and rotational speed on MBD biosorption at room temperature. The optimal parameters for MBD removal by Activated Carbon, Palm leaf, and Pomegranate peel were determined to be pH 5 and 1 g/100 ml dye solution, 120 min contact time for MBD concentration of 100 mg/l, and agitation speed of 250 rpm. The experimental data were analyzed using Langmuir and Freundlich isotherm models. The Langmuir biosorption isotherm was found to be the best model for simulation of MBD bisorption into Activated Carbon, Palm leaf and Pomegranate peel , with the Langmuir constant linked to the biosorption ability (q_{max}) of 7.87, 2.78 and 2.33 mg/g and a coefficient of determination (R^2) of 0.9996, 0.9985 and 0.9999 for Activated Carbon, Palm leaf and Pomegranate pee. The results indicate that activated carbon is the best environmentally friendly bio-adsorbent used in this study, which can be used efficiently to remove methylene blue dye from aqueous solutions.

Key word: Eco-friendly biosorbents, Methylene blue dye, Batch flow experiments, Isotherm models.

1 Introduction

Clean and fresh water is essential and vital as it sustains all forms of life on the earth, both as universal solvent and being a necessary for metabolic process of all organisms. So, the world's supply of clean water is running and the contaminant limits were established with getting stringent, to challenge all wastewater sources to meet these limits to provide and ensure clean water as the population increase[1]. Wastewater is used water generated from different activities related to public or private institutions. Dyes pollution in water resources is occurred when dyes pollutants is presented which are toxic contaminants in aquatic environment and is increased with the rapid development of industries, which is the main problem of water pollution. So that, it is necessary to check the water quality when even just 1 mg/l of dye concentration because of their toxic effects and carcinogenic to human and aquatic organisms, resulted some health problems. So, the removal of color from colored effluents becomes environmentally essential even a low dye concentration. Therefore, there is a constant need to effectively treatment methods which can effectively elimination these dyes[2].

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Dye removal technologies can be divided into three categories: chemical, physical, and biological. Membrane-filtration operations (reverse osmosis, nanofiltration, electrodialysis) and adsorption technologies are among the physical approaches. However, chemical approaches include coagulation or flocculation with flotation and filtration, electroflotation, precipitation-flocculation, traditional oxidation, electrokinetic coagulation, and electrochemical or irradiation activities. Biological methods comprise microbial degradation by fungal decolorization, adsorption by living or dead microbial, biomass and other bioremediation methods. Among these technologies, adsorption is found to be attractive due to it is cheaper, simpler, more efficient and economical than others. Thus, various adsorbents have been discovered and utilized to eliminate dyes in wastewater.

Various techniques for removing organic contaminants from wastewater have been investigated and implemented [3]& [4]& [5]. Where adsorption by solid adsorbents has the benefits of high efficiency, ease of design, and minimal start-up costs [6].

Nowadays, using agricultural biomass wastes as [Activated Carbon, Palm leaf and Pomegranate peel]an biosorbent has been adopted as the most feasible and affordable option for dyes removal. Using of natural materials like plant has been increasing attention due to their properties such as presence of constituents like pectin, protein, strulline, cellulose, lipid, and so on which help in biosorption due to their functional groups that act as binding agents and easily available, non-toxicity and being low cost alternative for dye removal

This work aimed to investigate the removal of Methylene blue dye (MBD) from polluted water by adsorption technique. Investigate the ability of prepared agricultural wastes as biosorbents for the MBD removal from aqueous solutions in batch. Examine the effects of several parameters on each biosorbent's biosorption capacity, such as dye solution pH, contact time, biosorbent dose, initial MBD concentration, and rotational speed.

2 Materials and Methods

2.1 Materials Bio sorbent

The biosorbents used in the present study was activated carbon produced from natural and local Iraqi plant(Schanginia/sp.), Palm leaf and Pomegranate pee,. These plant present in the different sites of Iraq.

2.1.1 Preparation of biosorbents

The activation of the appropriate plant material by carbonization was used to make activated carbon, Palm leaf, and Pomegranate peel. The raw material was cut into small pieces, and carbonization was carried out for one hour in a muffle furnace at 250°C. Based on earlier testing of various heating durations, this heating degree and time were determined. The plant was entirely carbonized to ash when the degree of heating and time were increased, according to the results of the several experimental trials. After carbonization, the carbon produced was rinsed with distilled water multiple times before being dried in an electric oven at 100°C to remove any unwanted moisture inside the particles and stored in desiccators. The biosorbent was then crushed and sieved through a 100 mesh sieve.

2.1.2 Adsorbate

All of the chemical reagents used in this study were analytical grade. Stock solution of MBD (1000mg/l) was prepared by dissolving certain amount of MB powder in distilled water and the

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desirable concentrations were prepared by diluting the stock solution. The residual concentration of MBD are measured using double beam UV-VIS spectrophotometer(UV/VIS-6800JENWAY) with maximum wave length. The maximum wavelength, λ max. for MBD was measured at 664. A calibration curve of MB concentrations versus absorbency was constructed as shown in Fig(1&2). The pH of dye solution was adjusted using Hydrochloric acid and sodium hydroxide with molarity of 0.1.

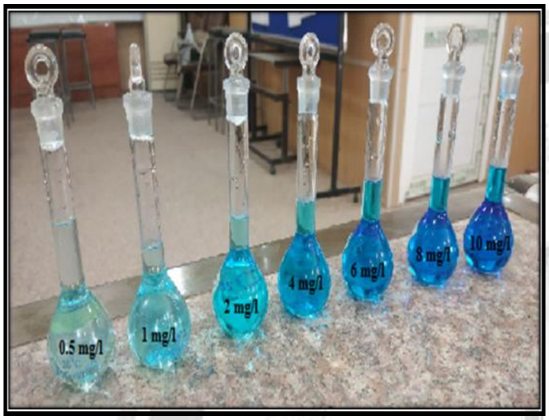

Fig.1: Different dye concentrations used for calibration curve.

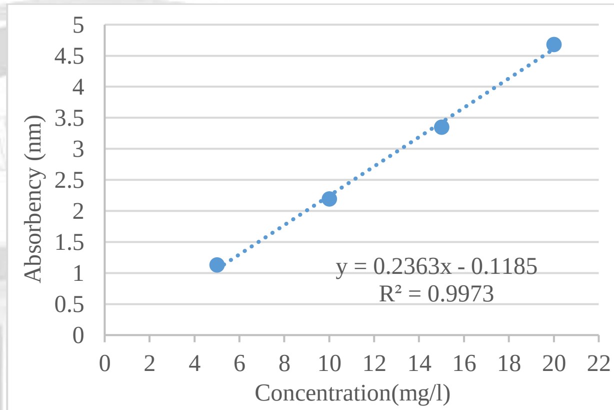

Fig.2: UV Spectrophotometer calibration curve of MB dye (at λmax=664 nm.)

2.2 Experimental Setup

Batch adsorption experiments

All batch trials took place at room temperature. The studies were carried out in a 250 mL series conical flask containing 100 mL of MBD solution and stirred with an average speed orbital shaker. Under various settings, pH, adsorbent dose, contact time, beginning dye concentration, rotational speed, and an adsorption isotherm research were investigated.

Biosorption isotherm studies

Biosorption isotherm studies were carried out by adding fixed amount of Carbon weight (1g) to different concentrations of MBD solutions (10-100 mg/l) in a series of 250 ml conical flasks filled with 100 ml at room temperature.

Then, the mixture was agitated for a predetermined period of time at a constant speed. The amount of dye adsorption at equilibrium qe (mg/g) was calculated from the following equation:

$$q_e = \frac{v}{w}(C_0 - C_e) \quad \ldots\ldots (1)$$

The definition of removal efficiency is as follows:

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

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Adsorption isotherm models

Langmuir isotherm

The Langmuir equation is frequently used to describe monolayer adsorption [7]. The model assumes a constant temperature, uniform adsorption energy, and a single layer of adsorbed solute.

$$q_e = \frac{q_m K_a C_e}{1 + K_a C_e} \quad \text{………………………………………………………………………..}(3)$$

It may be represented in the linear form as follows:

$$\frac{C_e}{q_e} = \frac{1}{q_m K_a} + \frac{1}{q_m} C_e \quad \text{…………………………………………………………………..}(4)$$

Where qe is the amount of MBD adsorbed at equilibrium time (mg/g), Ce is the concentration of MBD solution at sorption equilibrium (mg/l), qm and ka are the Langmuir constants corresponding to the maximum adsorption capacity (mg/g) and adsorption equilibrium constant (l/mg).

Freundlich isotherm

The Freundlich isotherm is an empirical equation employed to describe the heterogeneous system[8].

The equation is given below:

$$q_e = K_F C_e^{1/n} \quad \text{……………………………………………………………………………..}(5)$$

The linearized form of the Freundlich isotherm is shown below[9]&[10]

$$\log q_e = \log K_F + 1/n \log C_e \quad \text{………………………………………………………………}(6)$$

K_F and n are the Freundlich constants indicative of the relative adsorption capacity of the biosorbent ((mg/g)(l/mg)1/n) and the intensity of the adsorption respectively[11].

The adsorption feasibility of the adsorbent is expressed by the dimensionless separation factor, RL, related to Langmuir model using Eq7[12].

$$R_L = \frac{1}{1 + K_L C_0} \quad \text{……………………………………………………………………………}(7)$$

Because Co is the most common MBD contaminant (mg.L-1). RL classifies the isotherm shape as favorable (0 RL1), unfavorable (RL>1), irreversible (RL=0), or linear (RL=1).

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3 Results and discussion

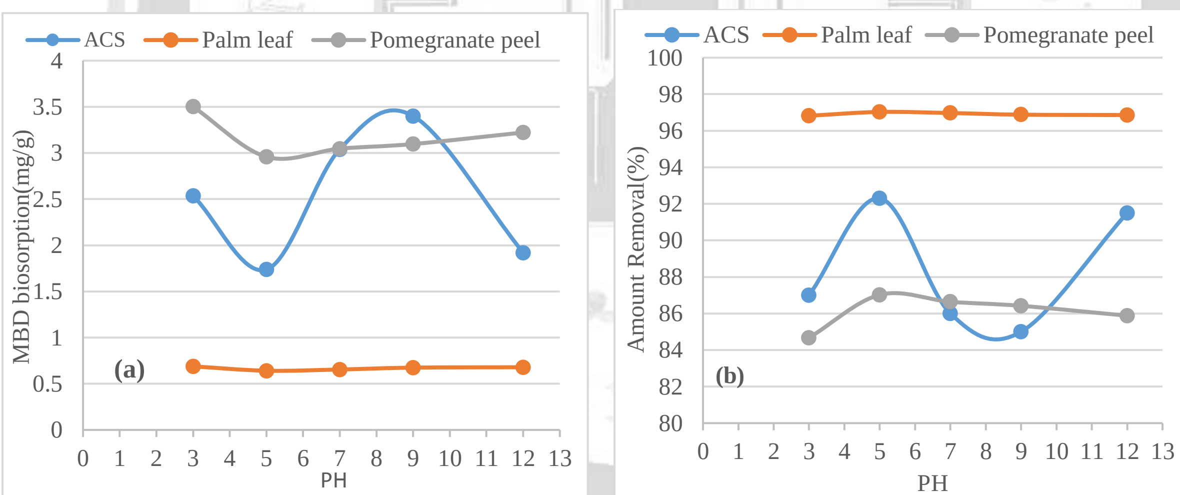
3.1 The effects of experimental parameters

❖ **Effect of solution pH**

The acidity of solution is the most important parameter controlling the adsorption process from aqueous solution, affects the functional groups on the surface of adsorbent, and define the solubility of contaminant in the aqueous solution, as well as affects the competition of contaminant ions that adsorbed to the binding sites of adsorbent [13].

The optimum removal of dye was reported at pH 5 and it was reported as 92.3%, 97.3% & 87.02% for Activated Carbon, Palm leaf and Pomegranate peel. Therefore, the next experiments were carried at pH 5, as shown in Fig (3).

The minimum biosorption of MBD onto eco-friendly biosorbents sorbent was reported at pH=3 because the more concentration and mobility of H_+ ions exist and the surface of sorbent is surrounded by hydrogen ions which compete with MBD cations for the biosorption sites of the biosorbents, preventing the dye ions from reaching the binding sites of the sorbents. When the pH value increased, the removal efficiency increased. This fact can be attributed to the weak inhibitory effect of hydrogen ions and increased negative charge intensity on the sorbent surface and the enhance the affinity of the positively charged MBD ions toward sorption sites[14]&[15].



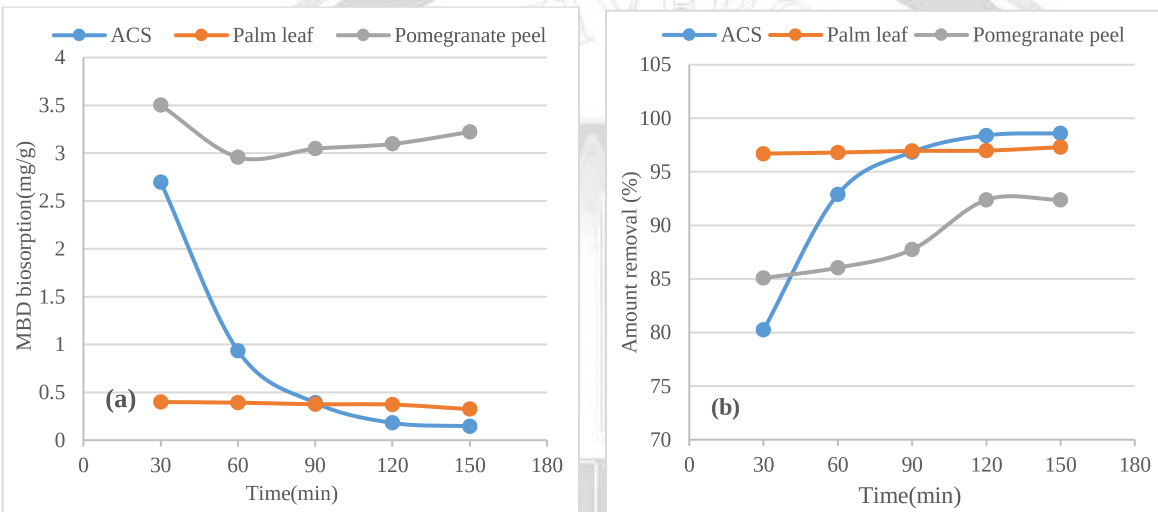
Fig(3):Effect of initial solution PH onto eco-friendly biosorbents

(a) MBD biosorption (b) MBD removal efficiency

❖ **Effect of contact time**

The research of the factor of contact time was prompted by the necessity to determine the ideal period for identifying the likelihood of dye binding for active sites occupied by dye and finishing its removal. The uptake and removal effectiveness of MBD by eco-friendly biosorbents as a function of contact duration in the range of 30-150 minutes is depicted in Fig(4).The maximum biosorption capacity for Activated Carbon, Palm leaf and Pomegranate peel was (2.698, 0.399 & 3.5018)mg/g.

From Fig (4), it can be seen that the biosorption process separated into two steps, fast and slow step. The first step included a rapid initial removal efficiency of MBD and it was over 80.25%, 96.7% &85.09% for Activated Carbon, Palm leaf and Pomegranate peel within the first 30 min. In the second step, a slow biosorption rate followed by reaching equilibrium at 120 min. This can be interpreted by the fact that at the beginning, a great number of surface sites are existing for biosorption of MBD ions and there is higher contact between the dye and surface of biosorbent, but after equilibrium time, the remaining surface sites are limited and difficult to occupy these ions[16]&[17].



Fig(4):Effect of contact time onto eco-friendly biosorbents

(a) MBD biosorption (b) MBD removal efficiency

❖ Impact of biosorbent dosage

The sorbent dosage is an important metric in determining the sorbent's capability for a particular initial sorbate concentration. Figure (5) shows the percentage of MBD removed using environmentally safe biosorbents.It can be shown that the percentage removal of MBD 99.13%, 97.5% & 92.38% using Activated Carbon, Palm leaf and Pomegranate peel with keeping other parameters (pH, contact time, MBD concentration and Rotational Speed)constant. This can be interpreted as the most MBD ions be sorbed onto sorbent and the equilibrium time was reached between MBD sorbed onto Activated Carbon, Palm leaf and Pomegranate peel . On the other hand, the amount of MBD sorbed per mass of biosorbent decreased frequently, because a great amount of sorbent reduced the unsaturation of the active sites, and after that the number of active sites per mass decreased, led a lower biosorption rate to a greater amount of biosorbent. Thus dose of 1g was chosen for biosorption of MBD using eco-friendly biosorbents in the next experiments[18].

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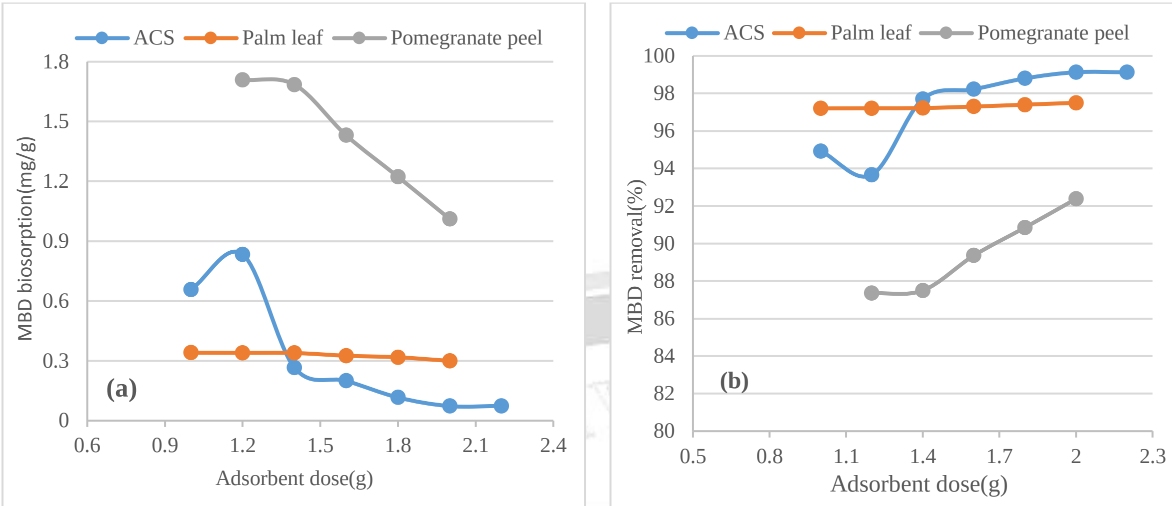

Fig(5): Impact of biosorbent dosage onto eco-friendly biosorbents

(a) MBD biosorption (b) MBD removal efficiency

❖ **Impact of primary MBD concentration**

Figure (6) depicts the absorption and percentage removal of MBD onto eco-friendly biosorbents as a function of initial MBD concentration in the range of 10-100 mg/l. The removal efficiency of Activated Carbon, Palm Leaf, and Pomegranate Peel reduced as the initial concentration of MBD increased.This is due to the fact that at a fixed dose of sorbent, a fixed biosorption area was available for MBD at higher initial dye concentration and a greater competitive occurs by the MBD ions remaining in the liquid media to be retained by the binding active sites of the sorbent. Equilibrium between the biosorbent dose and the sorbate concentration must be achieved the best conditions for efficient removal of sorbate. The biosorption capacity increased from (0.4933 to 3.658)mg/g for Activated Carbon , (0.2254 to 0.8907)mg/g for Palm leaf and (0.8664 to 2.9586)mg/g for Pomegranate peel. when the initial dye concentration grew from ten to one hundred milligrams per liter Two primary parameters may be used to interpret these results: a high rate of MBD diffusing onto the biosorbent surface and a high chance of MBD ions colliding with the surface of eco-friendly biosorbents [19].

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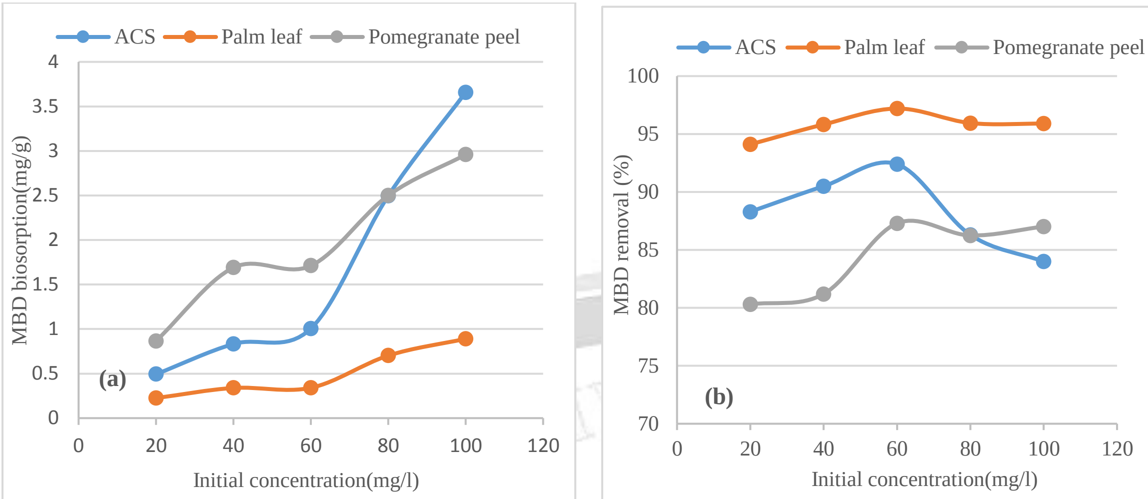
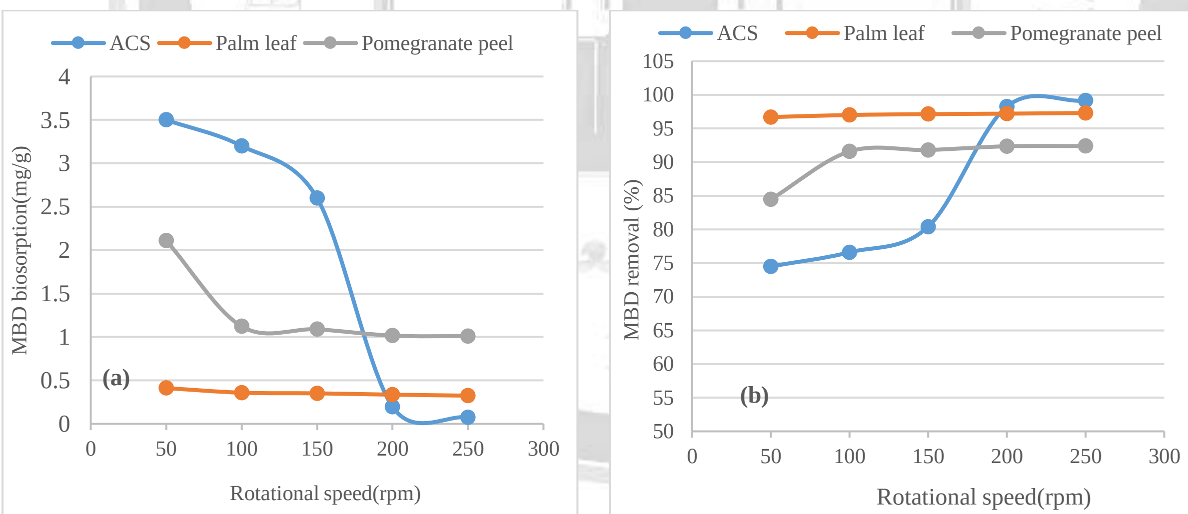


Fig(6): Impact of primary MBD concentration onto eco-friendly biosorbents

(a) MBD biosorption (b) MBD removal efficiency

❖ Impact of Rotational Speed

The optimum removal of dye was reported rotational speed at 250rpm and it was reported as 99.12 % , 97.3% & 92.39 % for Activated Carbon, Palm leaf and Pomegranate peel. Therefore, the experiments were carried at 250rpm as shown in Fig(7).



Fig(7): Impact of Rotational Speed onto eco-friendly biosorbents

(a) MBD biosorption (b) MBD removal efficiency

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3.2 Adsorption isotherm models

Langmuir isotherm

The Langmuir equation is frequently used to describe monolayer adsorption. The model assumes a constant temperature, uniform adsorption energy, and a single layer of adsorbed solute [20].

Isotherm analysis:

The adsorption isotherm is a critical component in the design of adsorption systems that rely on equilibrium data. The equilibrium data for MBD onto eco-friendly biosorbents [Activated Carbon, Palm Leaf, and Pomegranate Peel] were modeled in this study using two isotherm models, Langmuir and Freundlich, which correspond to the linearized forms Eqs (5 & 7). Figures (8,9, and 10) illustrate the plots of various models for biosorption.

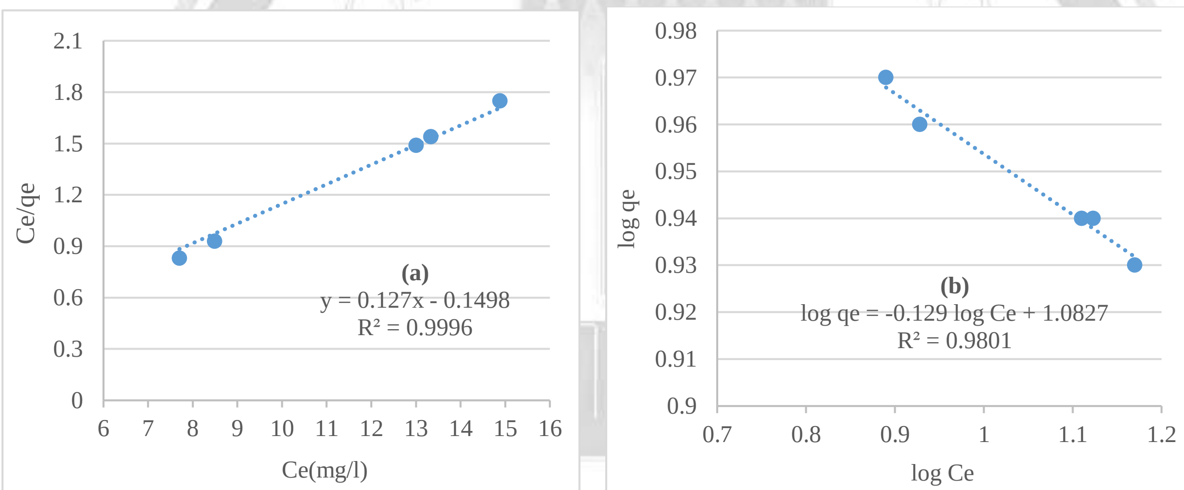

Fig (8): Linearized biosorption isotherm model of MBD onto Activated Carbon (a)Langmuir model (b)Freundlich model

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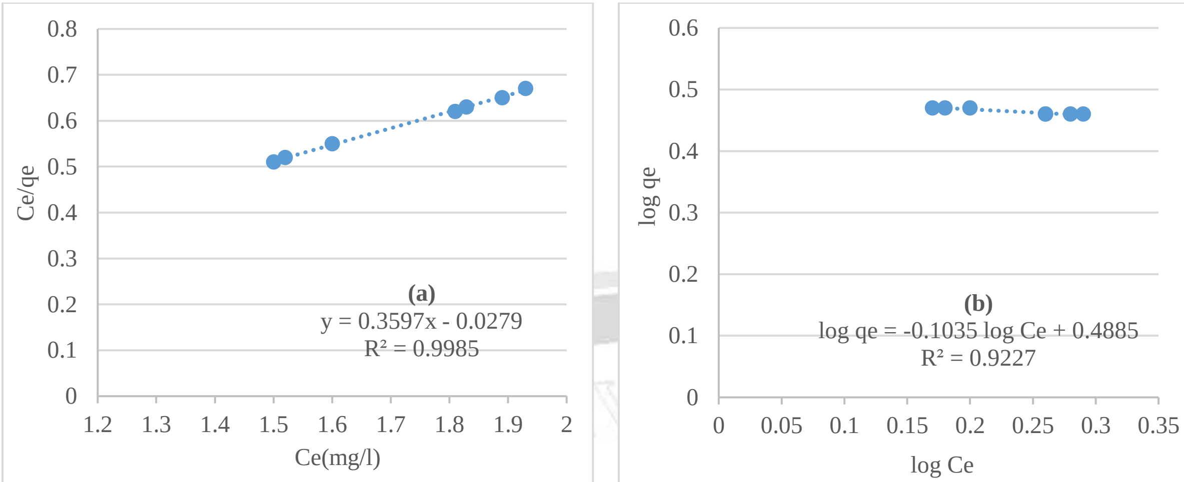
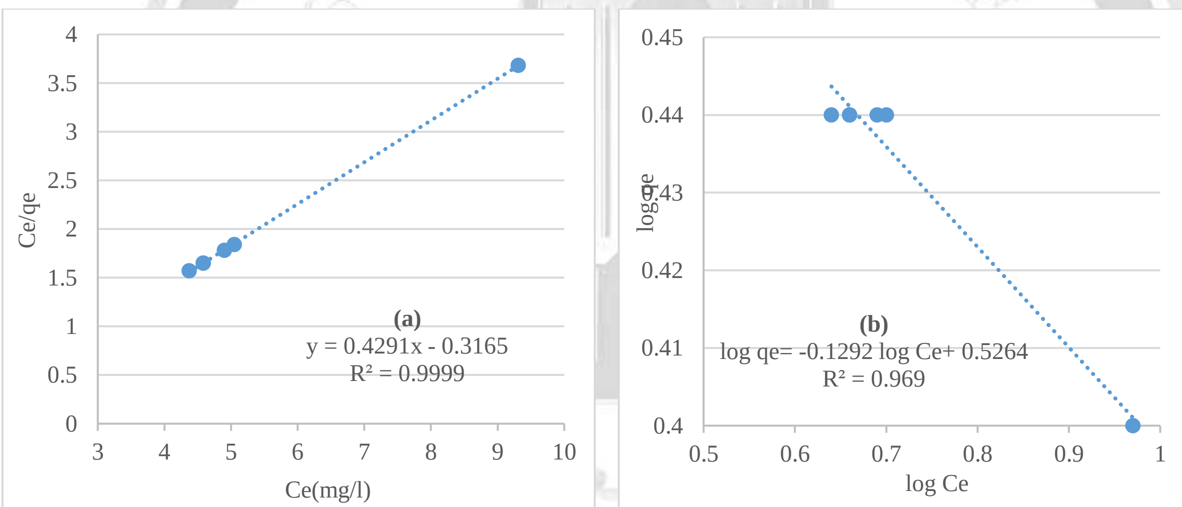


Fig(9):Linearized biosorption isotherm model of MBD onto Palm leaf (a)Langmuir model (b)Freundlich model



Fig(10):Linearized biosorption isotherm model of MBD onto Pomegranate peel (a)Langmuir model (b)Freundlich model

Tables (1) provide the calculated constants for each isotherm model with the determination coefficient (R^2) for MBD biosorption onto eco-friendly biosorbent.

Table (1): Constants of biosorption isotherm for MBD using eco-friendly biosorbent.

	Langmuir (95% Confidence level)		Freundlich (95% Confidence level)	
Activated Carbon	q_m(mg/g)	7.87	KF(mg/g)(1/mg)$^{1/n}$	12.09
	Ka(1/mg)	0.8	1/n	0.12
	R^2	0.9996	R^2	0.9801
	RL	(0.0123-0.0588)	Equation	

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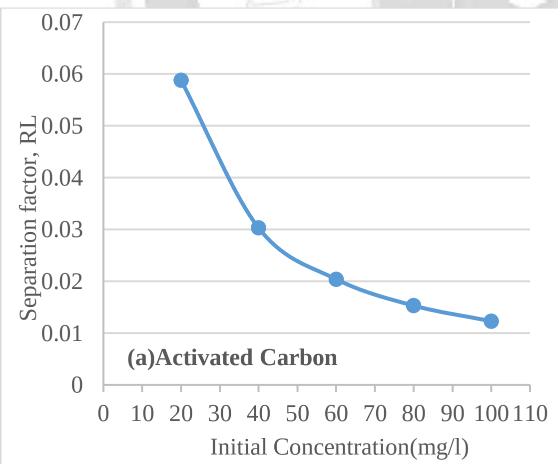
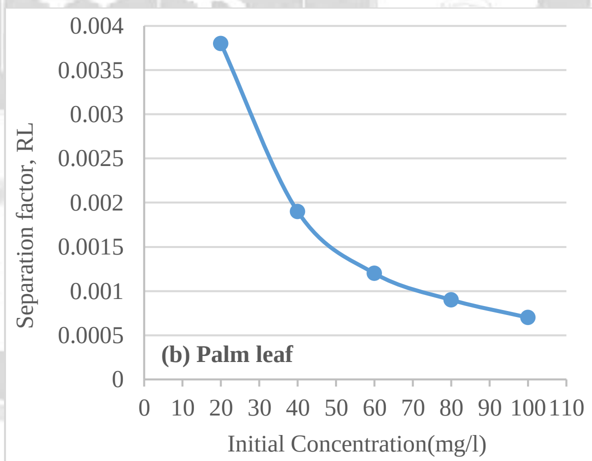
	Equation	qe=6.29Ce/1+0.8Ce		qe= 12.09 $Ce^{0.12}$
Palm leaf	q_m(mg/g)	2.78	KF(mg/g)$(1/mg)^{1/n}$	3.07
	Ka(1/mg)	12.9	1/n	0.10
	R^2	0.9985	R^2	0.9227
	RL	(0.0007-0.0038)	Equation	qe= 3.07 $Ce^{0.10}$
	Equation	qe=35.88Ce/1+12.9Ce		
Pomegranate peel	q_m(mg/g)	2.33	KF(mg/g)$(1/mg)^{1/n}$	3.34
	Ka(1/mg)	1.3	1/n	0.12
	R^2	0.9999	R^2	0.9690
	RL	(0.0076-0.0370)	Equation	qe= 3.34 $Ce^{0.12}$
	Equation	qe=3.02Ce/1+1.3Ce		

The maximum biosorption capacity (qm) was 7.87, 2.78 and 2.33 mg for MBD per gram of Activated Carbon, Palm leaf and Pomegranate peel respectively according to Langmuir isotherm model. The best fit of the data with the Langmuir isotherm for MBD indicated the uniform distribution of active sites surface on the sorbent . The value (Eq.7)for dye using eco-friendly biosorbent is cleared in Fig(11). The biosorption process of dye onto biosorbents was favorable when the values of RL were reported to be in the range of 0-1.





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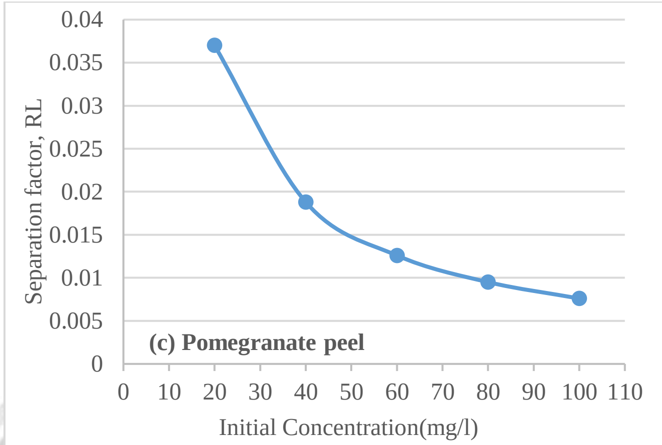

Fig(11):Values of separation factor,RL for biosorption of MBD onto eco-friendly biosorbent.

The value of the Freundlich constant (1/n), which was a measure of sorption intensity or surface heterogeneity, is becoming increasingly heterogeneous as its value approaches zero, according to the Freundlich isotherm model.As well as, the constant value (n) is between 1 and 10 also confirmed that the sorption was favorable and is beneficial as a low-cost biosorbent. Thus, the magnitude of the Freundlich constant indicates the easy sorption capacity of MBD from an aqueous media.

According to the Langmuir model, Activated Carbon has a maximum potential of 7.87 $mg.g^{-1}$ for the removal of MBD, which is higher than many other recorded biosorbents Table (2). Activated Carbon, on the other hand, may be called a very good low-cost biosorbent for the removal of MBD since it is a natural biosorbent that has not been processed or chemically changed.

Table (2): Comparison of maximum biosorption capacity (q_{max}) of eco-friendly biosorbent with other biosorbents reported for removal of MBD.

Adsorbent	q_{max} ($mg.g^{-1}$)	Reference
Alginate grafted polyacrylonitrile beads	3.51	Salisua et al., 2015
NaOH-modified rejected tea	2.44	Nasuha and Hameed, 2011
Defattedalgal biomass	7.8	Guarín et al., 2018
Water hyacinth root	2.89	Nibret et al., 2019
Activated Carbon	7.87	The present study
Palm leaf	2.78	
Pomegranate peel	2.33	

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4 Conclusions

1. Naturally occurring plant based Activated Carbon, Palm leaf and Pomegranate peel seems to be an effective and an alternative biosorbent for the removal of MBD from aqueous solutions.

2.Various operational factors were studied and the percentage dye removal increased as the pH of the solution and biosorbent dosage increased and the optimal value was reported at pH 5 and 1 g/100ml of dye solution respectively biosorbent dosage increased, but it decreased as the initial dye concentration increased

3. The biosorption of MBD was reached to equilibrium time at 120 min.

4. The Langmuir isotherm model was well-fitted to the Activated Carbon, Palm leaf and Pomegranate peel equilibrium biosorption of MBD, resulting in a homogeneous monolayer coverage of MBD on the Activated Carbon, Palm leaf and Pomegranate peel surface. The maximum biosorption potential was stated to be 7.87, 2.78 and 2.33 mg/g respectivily. Also, the dimensionless separation factor (RL) indicates the favourable biosorption process.

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ARTICLE
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مقارنة بين نتائج تطبيقات نموذجي لا نكمير و فراندلش لازالة صبغة الميثيلين الزرقاء من مياه الصرف الصحي الملونة

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الخلاصة

تتحرى هذه الدراسة عن قدرة المازات الحيوية الصديقة للبيئة منخفضة التكلفة، كمواد مازة لإزالة صبغة الميثيلين الزرقاء (MBD) من مياه الفضلات. أجريت سلسلة من التجارب المختبرية بنظام الدفعات لتقييم معالجة المحاليل المائية الصناعية باستخدام الكربون المنشط، ليف النخيل وقشر الرمان. تمت دراسة العوامل المختلفة التي تؤثر على عملية امتزاز الصبغة في درجة حرارة الغرفة بنظام الدفعات مثل الدالة الحامضية لمحلول الصبغة، وقت التلامس، جرعة المادة المازة، تركيز الصبغة الاولي وسرعة الدوران. تم التوصل الى ان درجة الحموضة ٥ وجرعة ١غم / ١٠٠مل من محلول الصبغة، و ١٢٠ دقيقة من وقت التلامس لتركيز الصبغة البالغ ٦٠ غم/لتر وسرعة الرج البالغة (250 rpm)، هي الظروف المثلى لإزالة الصبغة باستخدام الكربون المنشط، ليف النخيل وقشر الرمان. تم تحليل البيانات التجريبية عند الاتزان باستخدام موديلي لانكمير وفراندلش ، ووجد ان البيانات توافق موديل لانكمير ليكون افضل موديل لمحاكاة امتزاز الصبغة على سطوح الكربون المنشط، ليف النخيل وقشر الرمان. تم إيجاد قيمة الامتزاز القصوى (q_{max}) والبالغة (٧,٨٧، ٢,٧٨ و ٢,٣٣) ومعامل تحديد (R^2) يبلغ (٠,٩٩٩٦، ٠,٩٩٨٥ و ٠,٩٩٩٩) للكربون المنشط ، ليف النخيل وقشر الرمان على التوالي. تشير النتائج الى ان الكربون المنشط هو أفضل المازات الحيوية الصديقة للبيئة المستخدمة في هذه الدراسة والتي يمكن استخدامها بكفاءة لإزالة صبغة الميثلين الزرقاء من المحاليل المائية.

الكلمات الدالة: المازات الحيوية الصديقة للبيئة، صبغة الميثيلين الزرقاء، تجارب تدفق الدفعات، نماذج متساويةالحرارة.

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