USING LOW COAST MATERIAL (SEA GRASSES) AS ADSORBENT FOR REMOVAL ERIOCHROM BLACK T DYE FROM AQUEOUS SOLTIONS

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Abstract: Eriochrom T Black is one of most indicators used in different applications, especially for chemical analysis it is a dye which mainly causes many problems from environmental media as a water aqueous solution. The adsorbents of activated carbon used in this study were prepared from seagrasses. The kinetic behaviour of the process was investigated. In order to study the adsorption efficiency, many parameters, such as contact time and the adsorbent ratio, were used. Langmuir and Frundlish adsorption isotherm models were tested for the adsorption process. The linear regression coefficient R2 was used to elucidate the best-fitting isotherm models. Adsorption kinetics fitted well with the first-order kinetic model. The obtained results indicated succufilly adsorption of the studied dye using sea grasses activated carbon.

Keywords: Removal, Eriochrom.T, Sea grasses, Aquouse solution

INTRODUCTION

Environmental contamination of synthetic dyes in surface water and groundwater has a significant environmental impact. Dye-containing waste effects are generated by many sources such as textile, paper, printing, pulp mills, food, cosmetics and leather industries [1-2] Among the sources, the textile industry produces about 1.5 108 m3 of discharge volume of synthetic dye annually bout 1.5 108 m3 of discharge volume of synthetic dye and 10,000 different types of dyes annually [3]. Dyes are colored compounds of synthetic origin, which are resistant to fading upon exposure to light, chemicals and water. In addition, dyes have high organic content and non-biodegradable and complex benzene structure [4]. The presence of dyes in solution causes environmental damage to aquatic organisms by blocking sunlight, retarding photosynthetic activity and disturbing the re-oxygenation capacity, which creates an anaerobic condition that limits aquatic plant growth. In addition, contaminated surface water and groundwater would make the water bodies unsuitable for other uses [5] and can cause carcinogenic and mutagenic effects [6]. Among the industrial dyes, about 70% of the worldwide market used by dyeing industries is azo dyes. A reactive azo dye contains one or more azo bonds acting as chromophores in the molecular structure [7]. In addition, it is the most group of organic dyes that are difficult to degrade even at low concentration due to its high resistance to light, heat, water, chemical and microbial attack. Hence, it is imperative to remove azo dyes from wastewater effluents before their discharge into water bodies. Currently, several treatment technologies are used in the removal of dyes from waste effluents, such as electrochemical treatment, coagulation, precipitation, solvent extraction, membrane filtration, and advanced oxidation process [8]. However, these methods have disadvantages, such as high energy consumption, incomplete ion removal, and production of toxic sludge and other waste products that require further treatment and disposal. Among the physicochemical methods, adsorption is considered one of the most popular, efficient and comparable low-cost processes due to its simplicity. Among the variety of adsorbents, activated carbon has been proven to be effective in removing pollutants from water and even in gaseous environments [9].

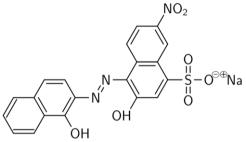
Activated carbon, a widely used adsorbent in industrial processes, comprises a micro porous, homogenous structure with a high surface area and shows radiation stability [10]. Its wide application is limited due to its high cost and difficulty in regeneration. Thus, research on producing activated carbon from renewable, low-cost Indigenous agricultural waste has gained attention worldwide because of its low cost and highly abundant characteristics [11].

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Some of the agricultural wastes that have been studied as possible sources of activated carbon are wood, bagasse, coir pith, orange peel [12], bamboo dust, corncobs, cassava peel, eucalyptus bark, palm shell, coconut shell, tobacco stem, hazel nut and some other biomass resources. Rice hull is an indigenous agricultural waste generated from the rice milling industry. It is regarded as non-valuable, and it is often dumped and burned, making it undesirable to the environment. On an annual basis, more than 140 million tons of rice hulls are produced, and 96% of this is contributed by developing countries [12]. Using rice hulls as adsorbent would reduce its disposal problems but also produce value-added products, such as activated carbon derived from rice hulls. However, using activated carbon from waste rice hulls has some disadvantages. Due to its high silica content, the thermally-activated adsorbents from rice husks exhibit low specific surface area. Several previous works on preparing activated carbon from rice hulls use common chemical activating agents such as ZnCl₂, H₃PO₄, KOH, NaOH and H₂SO₄ [13].

Activated carbon derived from rice husks has been utilized in the removal of heavy metals like cobalt, copper and lead [14], and various textile dyes such as Basic Blue and Reactive Orange [15]; CI Acid Blue 40 and CI Basic Blue 41; Acid Yellow; and Acid Blue [16 - 19]. Furthermore, there are only a few studies on removing Eriochrome Black T dye using activated carbon as an adsorbent. This study investigates the removal of Eriochrome Black T (EBT) dye from an aqueous solution using activated carbon (RHAC).

It is a complexometric indicator used in complexometric titrations, e.g. in the water hardness determination process. It is an azo dye. Eriochrome is a trademark of Huntsman Petrochemical, LLC.[20] In its deprotonated form, Eriochrome Black T is blue. It turns red when it forms a complex with calcium, magnesium, or other metal ions.



When used as an indicator in an EDTA titration, the characteristic blue end-point is reached when sufficient EDTA is added, and the metal ions bound to the indicator are chelated by EDTA, leaving the free indicator molecule. Eriochrome Black T has also been used to detect the presence of rare earth metals.[20] Sodium 4-[2-(1-hydroxynaphthalen-2-yl) hydrazin-1-ylidene]-7-nitro-3-oxo-3, dihydronaphthalene-1-sulfonate. The activated carbon of sea grasses as low-cost material to remove some dyes including two types of indicatore dyes including two types of indicatore dyes including two indicator types was used. The aims of this study can be summarized in the following points: Using the residual sea grasses to remove some industrial organic dyes such as chemical indicators including Eriochrom e Black T from aqueous solutions. Studying the impact of some of the factors including different dosages, different concentrations and effect of time of adsorbed materials. Using some mathematical relationships by calculating the Langmuir, Freundlich isotherms and adsorption kinetics.

EXPERIMENTAL

All chemicals used in this study are grade: The Eriochrom Black T from BDH company, different apparatus of shaker. Spectrophotometer (Type DU 800- Beckman coulter). Oven 30- 1000°C. Digital pH meter. Digital Balance, Digital Heater with thermostat system. The adsorption process was carried out according to methods reported by different studies [21 -25].

Preparation of activated carbon from residual sea grasses

Sea grasses were collected from the coast of Al-Hamama town (Libya). The sea grasses were washed several times with distilled water. They were dried overnight for several days, ground into a powder, and burned in the oven at 600°C for about three hours.

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Preparation of Eriochrome Black T Solutions

A stock solutions of the studied dyes of 1000 ppm were prepared by dissolving the appropriate amount of dye in water and made up to 100 ml mark with deionized water. Different concentrations of 100- 500 mg L⁻¹ of the dye were prepared from the stock solution. Deionized water was used for prepare all of the solutions and the reagents. A calibration curve of absorbance versus concentration was constructed using aUV-VIS spectrophotometer (Type D-U 800) at a maximum wavelength of 519 nm. Figure (1). The linearity equation was also obtained from standard curve Figure (2).

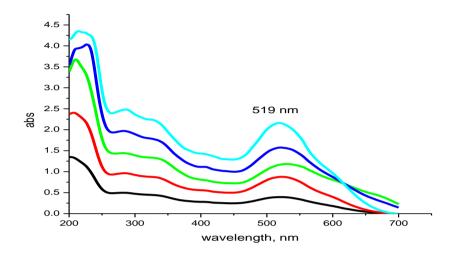


Figure 1: λ_{max} of Eriochrome Black T.

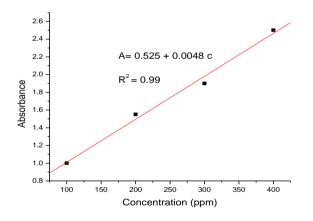


Figure 2. Standard Curve of Eriochrom Black T.

Adsorption factors.

Effect of Dosage.

Adsorbent dosage was optimized by performing the experiments at varying adsorbent dosages of (0.01, 0.02, 0.03, 0.04, and 0.05 g), with 10 ml of dye solution concentration 0f 100 mg/L concentration for Eriochrome black T, The bottles were shacked both for 20 min at room temperature then filtered. The absorbance of the dye solution was recorded by UV-VIS spectrophotometer.

Effect of Time.

To establish the effect of time on the absorption, the equilibrium investigations were carried out at the initial concentration of each reagent of 100 mg/L after selecting the best weight of adsorbent, which gave the high

percentage of removal (0.01 - 0.05 g) for dye. The adsorbent dose was added at different time of (5, 10, 15 and 20 min). The data obtained were used to plot the isotherm values.

Effect of concentrations.

The effect of the initial concentration on the removal of dye by adsorbents at a higher dose of each adsorbent was obtained. Experiments were carried out with a constant dose of the adsorbents, which showed high removal percentages.

Effect the temperature.

The Effect of temperature on the removal of selected dyes was studied at each concentration, where different temperature values ranged between (20 and 30 °C) of at fixed values of adsorbent dose and time.

Adsorption Studies.

Calculations of the capacity of adsorption.

The amount of dye adsorbed per gm (qe) was calculated based on the following equation:

$$(q_{\rm e}) = \frac{(c_{\circ} - c_{\rm e})}{m} \times v$$

 C_o and C_e are the initial and equilibrium concentration of adsorbate (here, Eriochrom Black T dye), respectively; V is the volume of dye solution (in litre); *m* is the weight of adsorbent.

The removal percentage of dye was calculated based on the following equation:

$$Removal \% = \frac{\boldsymbol{c}_{\circ} - \boldsymbol{c}_{\mathrm{e}}}{\boldsymbol{c}_{\circ}} \times 100$$

Adsorption Isotherms.

Adsorption isotherms can be generated based on theoretical principles. In order, two isotherm equations have been tested in the present research, namely, Langmuir and Freundlich, to describe the equilibrium characteristics of adsorption.

Langmuir adsorption isotherm.

The Langmuir equation is the most widely used isotherm equation for modeling the equilibrium.

The Langmuir linear equation is commonly expressed as follows:

$$\frac{Ce}{qe} = \frac{1}{kl} + \left(\frac{al}{kl}\right)Ce$$

A plot of Ce versus Ce/qe was linear, showing the applicability of Langmuir adsorption isotherm for Dye adsorption.

 K_L and a_L are the Langmuir constants related to adsorption capacity and rate of adsorption, respectively, which are calculated from the slope and intercept of the plot *Ce* versus *Ce/qe*.

The essential characteristics of Langmuir adsorption isotherm can be expressed in terms of a dimensionless constant, separation factor or equilibrium parameter 'RL', which is defined by,

$$R = \frac{1}{1 + al.Ci}$$

Ci = initial concentration of the dye and al = Langmuir constant. RL >1 Unfavorable, RL=1 Linear, 0< RL <1 Favorable, RL=0 Irreversible.

Freundlich adsorption isotherm.

The Freundlich isotherm model is the earliest known equation describing the adsorption process. It is an empirical equation and can be used for non-ideal sorption that involves heterogeneous adsorption. It also assumes that the adsorbent has a heterogeneous surface composed of adsorption sites with different adsorption potentials. This equation assumes that each class of adsorption site adsorbs molecules, as in the Langmuir equation. It is given by the following nonlinear equation below:

q=KC

KF is a system constant, related to the bonding energy. KF can be defined as the adsorption or distribution coefficient and represents the quantity of dye adsorbed onto the adsorbent for unit equilibrium concentration. 1/n indicates the adsorption intensity of dye onto the adsorbent or surface heterogeneity, becoming more heterogeneous as its value gets closer to zero. A value of 1/n below 1 indicates a normal Freundlich isotherm, while 1/n above 1 indicates cooperative adsorption.

The above equation can be linearized in the logarithmic form of the following equation, and the Freundlich constants can be determined:

$$logqe = logK_F + \frac{1}{n} logCe$$

A plot of log Ce versus log qe was linear, where kF measures adsorption capacity (mg/g), and n is adsorption intensity. 1/n values indicate the type of isotherm to be irreversible (1/n = 0), favorable (0 < 1/n < 1), unfavorable (1/n > 1). The values of 1/n and kF can be calculated from the slope and intercept, respectively.

RESULTS AND DISCUSSION

The results of the study parameters on the adsorbent of the studied dyes were described as follows.

Effect of dosage on the adsorption of Eriochrome Black T.

According to the results of removal percentage (%) of the effect of doses on the adsorption of Eriocheome Black T, showed that the dose of 0.03 g of sea grasses carbon gave the highest removal percentage value of (89.3 %) followed by the dose of 0.02 g (82.90%). On the other side, the lowest removal percentage values of (44%) were recorded for doses of (0.01g), respectively, as shown in Table (1) and Figures (3&4).

| Table1. | Effect of adsorbent | doses on the | adsorption | of Eriochrome | Black T at roo | om Temperature. |
|---------|---------------------|--------------|------------|---------------|----------------|-----------------|
| | | | | | | |

| Dose(g) | Final concentration (C _e)ppm | Removal % |
|---------|------------------------------------------|-----------|
| 0.01 | 55.90 | 44.10 |
| 0.02 | 17.10 | 82.90 |
| 0.03 | 16.65 | 89.35 |
| 0.04 | 19.35 | 80.65 |
| 0.05 | 21.67 | 78.33 |

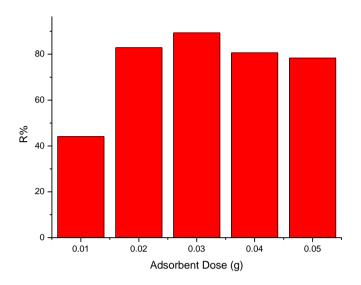


Figure 3. Effect of adsorbent doses on the adsorption of Eriochrome Black T at (25°C).

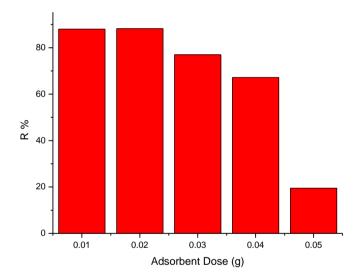


Figure 4. Effect of adsorbent doses on the adsorption of Eriochrome Black T at (30 °C).

This indicates that at the adsorbant's contents, the adsorption of Eriocheome Black T (Removal percent) gave a high rate compared with the other high doses(g). This is important for the used activated carbon whichimportant for the activated carbon obtained from sea grasses, because small quantities from low low-cost materials gave high percentages of removal of the Eriochrome Black T.

Effect the time on the adsorption of dyes.

From the results shown in Table (2), there are variations of the applied time (min) on the removal percentage of the Eriochrome Black T, where the high removal percentage was obtained at (20 min), the results also indicated that the percentage of removal the dye was increased with increasing the applied time.

| Time (min) | q _e (mg/g) | Final Concentration C _e (ppm) | C _e /q _e |
|---------------|-----------------------|---------------------------------------------|--------------------------------|
| 0 | 5.80 | 0 | 0 |
| 5 | 6.79 | 17.10 | 2.85 |
| 10 | 6.28 | 16.65 | 2.65 |
| 15 | 4.86 | 19.35 | 2.61 |
| 20 | 8.06 | 21.67 | 2.60 |

| Table 2. Effect of time on the adsorption of ErioChrome Black T. |
|------------------------------------------------------------------|
|------------------------------------------------------------------|

Adsorption Isotherms (Langmuir and Frindulich).

Langmuir isotherms.

The most widely used isotherm equation for modelling the equilibrium is the Langmuir equation. The Langmuir linear equation is commonly expressed as follows:

$$\frac{Ce}{qe} = \frac{1}{kl} + \left(\frac{al}{kl}\right)Ce$$

A plot of Ce versus Ce/qe was linear, showing the applicability of Langmuir adsorption isotherm for bromo cresol purple and cresol red adsorption.

 K_L and a_L are the Langmuir constants related to adsorption capacity and rate of adsorption, respectively, which are calculated from the slope and intercept of the plot *Ce* versus *Ce/qe*. The effect of different times of adsorbent on the adsorption of the studied dyes was monitored. A linearized of C_e versus C_e/q_e was obtained, The fits are a guite for the sorbent to suggest applying the Langmuir model for the investigated system. The values of Langmuir parameter with correlation were computed from the intercept and the slope of the fitted Langmuir equation, were the values of Al ,KI and r2 were 0.28233, 0.357 and 0.92, respectively.

Both values of r^2 of the studied dyes seem high enough to assign the successful application to adsorbent the dyes on the sea grasses. The essential characteristics of a Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor RL as follows.

$$\mathrm{RL} = \frac{1}{1 + al \cdot Ci}$$

Where, Ci = initial concentration of the dye and al=Langmuir constant.

The equilibrium isotherms types are related to the RL values for RL > 1 Unfavorable, RL = 1 Linear, 0 < RL < 1Favorable, RL=0 Irreversible. In the present study, the values of RL were found to be less than (1) and slightly higher than (0) value indicating the favorable adsorption of the selected dyes on the sea grasses, In this study, the **RL** of Eriochrome Black T was (0.263)

Freundlich adsorption isotherm.

The Freundlich isotherm model is the earliest known equation describing the adsorption process. It is an empirical equation and can be used for non-ideal sorption that involves heterogeneous adsorption. It also assumes that the adsorbent has a heterogeneous surface composed of adsorption sites with different adsorption potentials. This equation assumes that each class of adsorption site adsorbs molecules, as in the

Langmuir equation. It is given by the following nonlinear equation below:

q=KC

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KF is a system constant related to the bonding energy. KF can be defined as the adsorption or distribution coefficient and represents the quantity of dye adsorbed onto the adsorbent for unit equilibrium concentration. 1/n indicates the adsorption intensity of dye onto the adsorbent or surface heterogeneity, becoming more heterogeneous as its value gets closer to zero. A value of 1/n below 1 indicates a normal Freundlich isotherm, while 1/n above 1 indicates cooperative adsorption. The above equation can be linearized in the logarithmic form of the following equation, and the Freundlich constants can be determined:

$$\log qe = \log K_F + \frac{1}{n} \log Ce$$

A plot of log Ce versus log qe was linear, where kF is a measure of adsorption capacity and (n) is adsorption intensity. 1/n values indicate the type of isotherm to be irreversible (1/n = 0), favorable (0 < 1/n < 1), unfavorable (1/n > 1). The values of 1/n and kF can be calculated from the slope and intercept, respectively, the values of Freundlish $(1/N, \text{KF} \text{ and } r^2)$ were , 0.6694, 2.522 and 0.99, respectively.

The results of Freundlich's present study were illustrated in Table (8) and represented in Figure of (3).

| Time (min) | Final Concentration C _e (ppm) | log C _e | q _e (mg/g) | Log q _e |
|---------------|------------------------------------------------|--------------------|-----------------------|--------------------|
| 0 | 0 | 0 | 0 | 0 |
| 5 | 17.10 | 1.23 | 6.09 | 0.784 |
| 10 | 16.65 | 1.22 | 6.28 | 0.797 |
| 15 | 19.35 | 1.28 | 7.41 | 0.869 |
| 20 | 21.67 | 1.33 | 8.33 | 0.92 |

Table 3. The values of Freundlich isotherm for E.B.T.

Kinetics of adsorption.

According to the values obtained from the isotherms in this study, for. The adsorption process follows the firstorder reaction. The Kinetics of the adsorption was conducted by the values recorded according to the Effect of time on adsorption. The values are shown Table (4) and Figure (5).

Table 4. Effect of time on the adsorption of Eriochrome Black T.

| | Time(min) | Concentration(ppm) |
|----|-----------|--------------------|
| 0 | | 0 |
| 5 | | 1.23 |
| 10 | | 1.22 |
| 15 | | 1.28 |
| 20 | | 1.33 |
| | | |

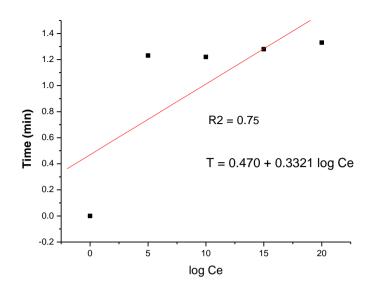


Figure 5. Effect of time on the adsorption of Eriochrome Black T.

The adsorption rates are measured by determining the dye concentration as a function of time. (C_t is the concentration of dyes at different times) versus time (min). The adsorption rates were calculated from the slopes the results recorded that the rate of Adsorption rate (K) of activated sea grasses was 0.331 which mainly indicated that the adsorption following first order rate.

CONCLUSION

This research aims to evaluate the efficiency of low-cost adsorbents of activated carbon obtained from sea grasses. The noteworthy observations and conclusions can be summarized as follows:

The adsorption efficiency of dyes onto activated carbon increases in contact time till they reach equilibrium. An increase in adsorbent dose led to % removal while a decrease in adsorption capacity. The adsorption isotherm studies showed that the best Langmuir isotherm was for the adsorption of Eriochrome Black T. However, the best Freundlich isotherm was recorded for adsorption of Dye.

Conflict of interest

There was no conflict of interest declared by the authors.

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