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Adsorption Studies of Cationic Dye on Raw And Modified Sugarcane Bagasse from Aqueous Solutions: Kinetic and Isotherm Aspects

Sara E. Abd Elhafez,[a*] Azza El-Maghraby,[a] Nahla A. Taha [b]

[a] *Department of Fabrication Technology, Advanced technology and New Materials Research Institute, City for* Scientific Research and Technology Applications, Alexandria, Egypt.

[b] *Department of Modeling and simulation, Advanced Technology and New Materials Research Institute, City of Scientific Research and Technological Applications, SRTA-City, Alexandria, Egypt.*

Abstract

Adsorbent materials for industrial and environmental applications are the goal of seeking research under active investigation now. Activated carbons could be obtained from the organic material after been chemically or physically treated. In Egypt, Sugarcane bagasse could be considered one of the agricultural wastes that frequently produced. In this study, the low-cost adsorbent was prepared from raw Bagasse. The batch experiment was set up to determine the effect of using the agriculture waste (sugarcane bagasse) without any modification and activated it chemically and physically on methylene blue removal. The influence of adsorbent amount, agitation rate, and temperature on the dye removal were studied. The result shows that the percentage of dye removal reached up to %99.6 when the initial dye concentration was 15 ppm, and the amount of sorbent (chemically activated) was 0.3 g at room temperature. The adsorption kinetics and isotherms for the removal of methylene blue were represented with different models. It was detected that Langmuir and Freundlich's equations have the R^2 value closed to unity, and the maximum adsorption calculated capacity was 2.2 mg/g when the initial dye concentration was 15 ppm. Furthermore, pseudo-first-order and pseudo-second-order were fitted to examine the adsorption kinetic, and a pseudo-second-order calculated the highest adsorption rate for the raw bagasse (0.59446 g/mg.min).

Keywords: Cationic dye; Methylene blue; Sugarcane bagasse; Kinetic; and Isotherm.

1. Introduction

Dyes are mainly organic complexes that can change the colour of material by attaching to its surface. The majorities of colours are organic matrix molecules that needed to be fixed and resisted to a lot of things such as they react with detergents. Synthetic dyes are widely used in many fields of advanced technology such as in various kinds of paper [1], food processing [2], printing, leather tanning, plastics, textile[3], and dye manufacturing industries [4].

The main problem in using dyes is its non-degradable nature, which makes its discharge severely effect on the environments[5].

Adsorption could be considered an affordable method of wastewater treatment, which able to minimize the dangerous pollution in the industrial effluents[5][6][7]. Several studies were conceded to reduce the adsorption process's cost by using the agricultural by-product as a cheap bio-sorbent [8]. Many researchers have focused on increasing the capacity of the sorbent by applying different treatments [9]. The adsorption process can be classified into two types: (1) chemical adsorption, (2) physical adsorption. Chemical adsorption occurred when strong bonds formed between the molecules or ions of adsorbent and adsorbate. Physical adsorption is a generally reversible process, whereas the week Vander weals bond bent between adsorbent and adsorbate. Adsorption process has a number of advantage such as affordable, sample equipment, flexibility design, less in toxic pollutants , reduce harmful effluence and, ease in operation. The main features that affect the efficiency of adsorption process include the attraction between the adsorbate-adsorbent, the effective area on the adsorbent surface, adsorbent weight, adsorbent particle size, contact time, temperature, and pH of the solution [10][1][8].

Activated carbon (AC), an amorphous phase of carbon, has been produced from treated material to built pore structure highly; therefore, effective adsorbent with high surface area to volume ration was formed[11] [12]. Many methods could be used to

*Corresponding author e-mail: shafez@srtacity.sci.eg.;.

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produce AC, such as chemical activation and physical activation. Nevertheless, activated carbon has many drawbacks, such as the preparation cost is high, low selectivity, and lacking effectiveness against dispersing and cask dyes. These reasons have encouraged scientists to search on cheaper source for activated carbon. The massive amount of agricultural by-product such as Theobroma cacao [13], rise straw [7], date stones/pits [14] , palm fibers [15], orange peel[16], pomegranate peels [3], and Peanut hull [17] make them one[18] of the critical starting material for activated carbon (AC) production. Activated carbon could be produced theoretically from any carbonaceous material with a considerable amount of elemental carbon[19] [5]

Bagasse could be considered one of the most significant agricultural waste products as it has produced annually in large quantities[15]. Sugarcane bagasse, which is composed of macromolecules, is a residue from the sugarcane crushing process. These macromolecules have hydroxyl groups which react chemically to form an active adsorbent material with accepted properties[20]. The usage of bagasse as adsorbent material to remove different pollution was illustrated in the literature [21].

This study aimed to determine the efficiency of used the sugarcane bagasse in its raw and activated carbon phase on the removal of the cationic dyes, methylene blue (MB), from Synthes dye solutions. The adsorption process was studied using several parameters: adsorbent weight, agitation rate, and temperature.with several initial dye concentrations. The results were analyzed by two kinetic models and two isothermal adsorption models.

2. Materials and methods

2.1. Material and apparatus

Methylene blue (MB) $C_{16}H_{18}N_3ClS$ (supplied by Nice Chemicals Pvt. Ltd) was used as the adsorbate. P\hosphoric acid (H3PO4) (85%) (Riedel-de Haen) was used to activate the bagasse. Sodium Chloride (NaCl) (Universa Laboratories PVT, Mumbai), Sodium Hydroxide (NaOH) (Central Drug House, New Delhi)) and Hydrochloric acid (37%) (Merck, U.S.A) was used as the reagent to evaluate the adsorbent surface charge. The synthesis solution was prepared using distilled water.

2.2. Preparation and characterization of activated carbon

Sugarcane bagasse was used in this study as a raw material for preparing the adsorbent obtained locally. Sugarcane bagasse was washed thoroughly with tap water to remove the adhered particles; then, it was put in the oven (Nabertherm, TR 120, Germany) at 70^0 C for dring till obtained constant weight. Then it was crushed to the desired mesh size (1–2 mm). The prepared raw material was separated into three portions. The first portion was activated physically. It was pyrolyzed at 600◦C in a steel vertical tubular reactor placed in a muffle furnace (Barnstead Thermolyne, F48010, U.S.A) for one hour [2]. The carbonized material was stored in desiccators and labelled as physically activated carbon (PC). The second portion was activated chemically. Sugarcane bagasse was subject to digestion with H_2PO_4 as let bagasse to boil in 40% H_2PO_4 for one hour (mass ratio of bagasse to phosphoric acid 1:9.8) by using a mechanical stirrer with heater. Then the mixture was filtrated and washed with distilled water to remove the remaining chemicals then let it dry in air. Treated material was put in a muffle furnace for carbonization at 500°C for 1 hour. Finally, The carbonized material is characterized as chemical activated carbon (CC) and stored in desiccators. The third portion was used as raw bagasse (RB) without any treatment. The characteristics of bagasse and its activated carbon were made in this study Scanning electron microscope (SEM) and further analysis has been discussed in our previous study[5].

2.3. Determination of the point of zero charges (PZC)

Determination the PZC of the raw adsorbent, pH value of the 25 mL of NaCl (0.1 M) was adjusted using (0.1 M) NaOH or HCl. pH value ranged approximately from 2 to 12. Then, 0.25 g of (RB) was added to the solution and shaken at 200 rpm for 48 h. After settling, samples were filtered, and the final solution pH values were measured. The pH of the pzc of the raw bagasse adsorbent was obtained from the intersection between Δ pH (the difference between final and initial pH) against the initial pH.

2.4. Batch equilibrium studies

The batch adsorption experimental was tested in a set of conical flasks (100 ml) where solutions of dye (50 ml) with different initial concentrations of cationic dye (PH=8.7) (10-30 ppm) were used in this study. Different weights of raw and its activated carbon were added to dye solutions and kept in the shaker at room temperature for one h (equilibrium time) of the solid-solution mixture[5].

The flasks were then brought out from the shaker, and its PH was measured for solution treated with raw bagasse (7.72) and the physically activated (7.88). The final concentration of dye in the solution was analyzed by estimated the absorbance of the supernatant solution with a double beam spectrophotometer (Ultropec 2000 UV/visible) at wavelength 665 nm. Each experiment was duplicated under identical conditions. The following equation calculated the amount of adsorption at equilibrium, q_e (mg g^{-1}), and the % dye removal (R):

$$q_e = (C_0 - C_e)\frac{V}{m} \quad \text{Eq. 1}$$

$$\%R = \frac{C_0 - C_e}{C_0} * 100 \quad \text{Eq. 2}$$

Where: C_0 and C_e are the initial and equilibrium dye concentrations in the liquid-phase, respectively (mg L^{-1}). V is the volume of the Solution (L), and m is the mass of dry adsorbent [g].

3. Results and Discussion

3.1. Characterizations of raw and activated bagasse

The SEM images of (RB), (PC) and (CC) are shown in Fig. 1. As shown in Fig. 3 (a) the hollow tube is main morphology of the raw materials. After further treatment shown in Fig. 3(b), the physically activated microparticles are, thereby increasing the adsorbent's active sites and surface area and enabling it to adsorb the MB dye further. Fig. 3(c) shows that the porous, rough surface of the chemically activated adsorbent has appeared beside the broken hollow tube shape surface. In particular, the treating which applied on the (RB) has radically enhanced the surface area that exposed to the adsorption processes.

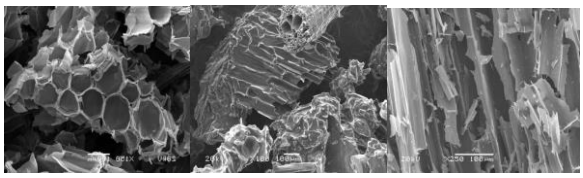

Fig. 1 SEM of peanut hulls surface with magnification factor (a) raw bagasse (RB) (b) physically activated carbon (PC) (c) chemical activated carbon (CC)

3.2. Determination of pzc

Dye removal by adsorbent is stated to be profoundly affected by solution pH value. The point of zero charge of raw adsorbent was found 6.3. As such, the charge on the bagasse surface was positive when the Solution pH lower than 6.3. by contrast, the charge on the bagasse surface was negative when the Solution pH higher than 6.3. from this, it could be concluded that the cationic dye (MB) is favourable in the pH above 6.3.

3.3. Effect of the adsorbent amount

Investigation the effect of the adsorbent amount of raw bagasse and two modified types on the removal of MB dye, different amount of adsorbent was used (from 0.05 to 0.5 g/50 ml) in the dye solution. The synthetic solutions were prepared with the different initial dye concentrations ranged from 10 to 30 ppm, and the contact time was 60 min.

Generally, the percentage of dye removal increases with increasing the adsorbent weight, as the number of sorption sites at the surface of adsorbent increase by increasing the amount of the adsorbent. Results in figures (2-4) showed that the adsorption capacity of MB dye in the first stage increase with the increase in the adsorbent dose to achieve approximately complete dye removal (99% removal) when using 0.3 g of the chemically treated bagasse with different dye concentration. Then it remained constant or decrease slightly with the further increase in the adsorbent dose due to the conglomeration of adsorbent particles, which led to no increase in active sites on the adsorbent site.

At low dye concentrations, the vacant surface sites adsorb the dye more rapidly. The percentage of dye removal when using the same amount of physically treated bagasse increased by 10% when the concentration decrease from 30 to 10 ppm. One explanation for this observation may be the fact that at low concentration, almost all dye molecules are adsorbed very quickly on the outer surface of the sorbent. Further increases in the initial dye concentration led to fast saturation of the adsorbent surface. Thus, most of the dye adsorption took place slowly inside the pores by intra-particle diffusion. Steric repulsion between molecules of the solute can also minimize the adsorption process. It was found that the 0.3 g of the adsorbent gave the highest dye removal (i.e., 99.6% and 89% for chemically and physically treated bagasse, respectively, while 0.1 gm was the optimum concentration when raw badass was tested.

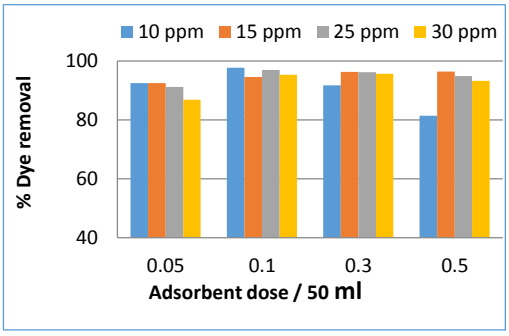

Fig. 2 The variation of dye removal percent with the adsorbent dose with different initial MB concentrations at room temperature for raw bagasse.

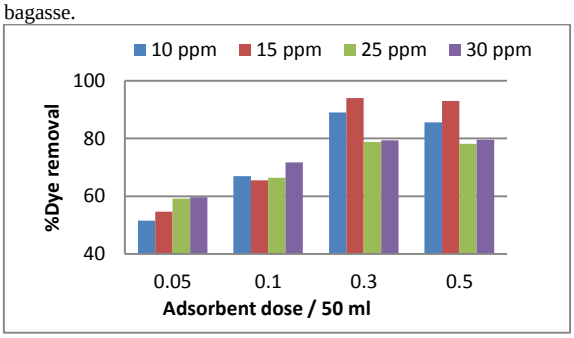

Fig. 3 The variation of dye removal percent with the adsorbent dose

with different initial MB concentrations at room temperature for physically activated carbon

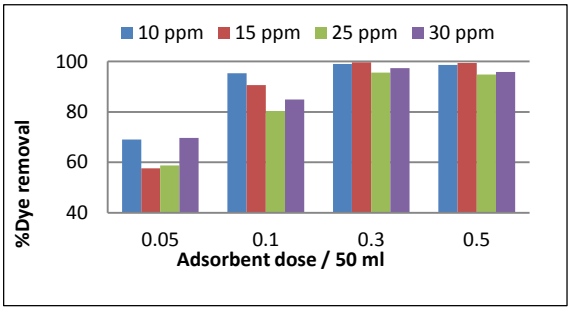

Fig. 4 The variation of dye removal percent with the adsorbent dose with different initial MB concentrations at room temperature for chemical activated carbon.

3.3. Effect *of* agitation rate

The agitation enhances the chance of adsorbate contact with the adsorbent. The influence of variable stirring rate on the dye removal efficiency of sugarcane bagasse and its modified form are depicted in figures (5-7). Exploration this effect on the removal of methylene blue dye, the stirring rate was varied from

10 to 400 rpm keeping the initial dye concentration ranged from 10 to 30 ppm, the contact time was 60 min at 25 ^{0}C, and the adsorbent dose was 0.3 g. It was observed that at the lower rate of stirring, the removal capacity was little, but as the rpm increased, the dye removal also increased. This increase might be due to the improved contact between the adsorbate and the adsorbent at higher rpm. In this study, the optimum stirring rate was considered as 100 rpm.

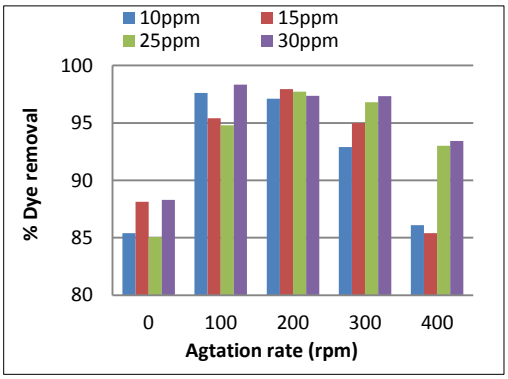

Fig. 5. The variation of dye removal percent with agitation rate with different initial MB concentrations at room temperature for raw bagasse

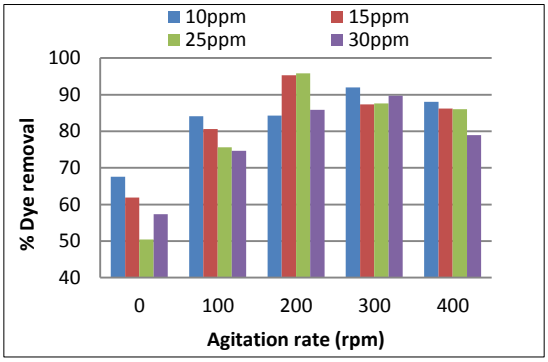

Fig. 6 The variation of dye removal percent with agitation rate with different initial MB concentrations at room temperature for physically activated carbon.

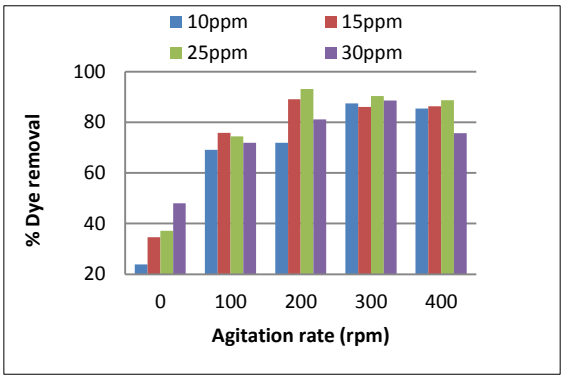

Fig. 7 The variation of %dye removal percent with agitation rate with different initial MB concentrations at room temperature for chemical.

3.4. *Effect of temperature*

The effect of temperature on the adsorption process was studied from 20 to 55 °C. An increase in temperature resulted from a very slight decrease in dye adsorption. At high temperature, the binding between adsorbent and adsorbate was weakened, which led to the detachment of the adsorbent. In this study, as shown in figures (8-10), it was found that the temperature is slightly affected by the dye removal by all studied adsorbent. It is preferable to operate at room temperature as the % dye removal reached 97.96%[22][23].

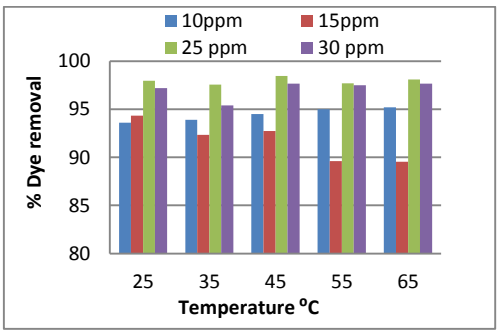

Fig. 8.The variation of %dye removal percent with the temperature at various initial MB concentrations for raw bagasse

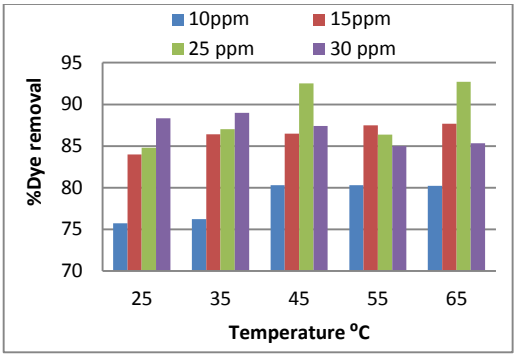

Fig. 9 The variation of % dye removal percent with the temperature at various initial MB concentration for physically modified raw bagasse

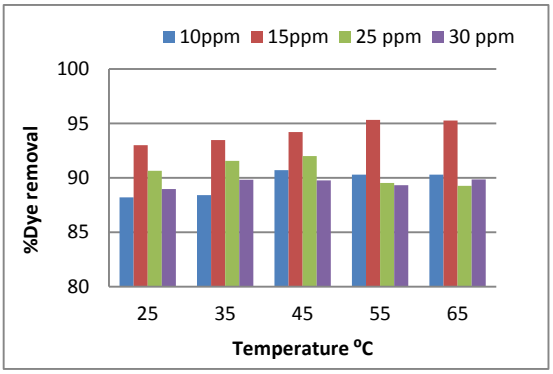

Fig. 10 The variation of %dye removal percent with the temperature at various initial MB concentration chemically modified raw bagasse.

3.5. Dye adsorption kinetic-study

The kinetic study for the adsorption process illustrates the controlled mechanism of adsorption process reaction rate-controlling mechanisms of adsorption process such as chemical reaction, diffusion control, or mass transfer coefficient are used to determine kinetic models. The study of adsorption kinetics illustrates how the solute uptake rate and obviously, this rate controls the residence time of the adsorbate at the solution interface. The determined reaction rate one of the essential parameters has taken when the full- sale batch process has designed.

The dye decolourization kinetics by the raw bagasse and its modified phases were studied for the methylene blue dye.

The original data for the dye concentration equal to 15 ppm was used for the linear plots of the adsorption kinetics. The linear plots of pseudo-first-order and pseudo-second-order at 303 K are presented in Fig. (11-12). The pseudo-first-order kinetic model with R2 values was generally more significant than 0.9970. However, the adsorption of methylene blue by raw bagasse best fit the pseudo-second-order model because the mean value of R2 for the initial methylene blue concentration tested was 1. The two plotted models were linear with R2 values generally closed to unity suggesting the applicability of both models to the kinetic data. The correlation coefficient and models' parameters are presented in table1.

Adsorption rate constants were determined using of pseudo-first-order Eq. 3 and pseudo-second-order Eq. 4 and Eq. 5 at 303 K

$$\ln(q_e - q_t) = \ln q_e - k_1 t \qquad \text{Eq. 3}$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad \text{Eq. 4}$$

$$h = k_2 q_e^2 \qquad \text{Eq. 5}$$

Where: q_t is the amount of dye adsorbed at time t (mg/g). t is the contact time (min), k_1: is the rate constant of the pseudo-first-order adsorption (min -1). k_2 is the rate of sorption (g/mg. min) h: is the initial sorption rate, (mg/g.min).

The chemically-treated bagasse exhibited the highest degradation rate among other studied adsorbents, showing a reaction rate constant of 0.6323 min−1 (R^2 = 0.9996).

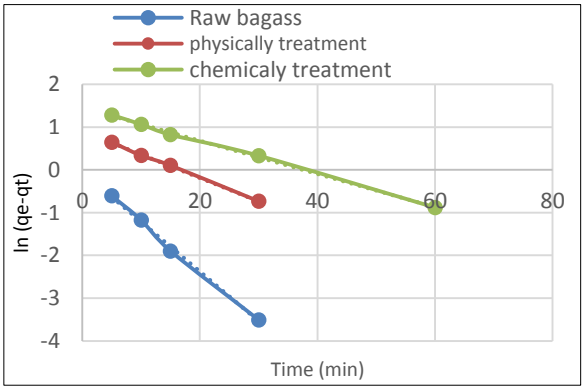

Fig. 11 Pseudo first-order kinetics plot for the adsorption of methylene blue dye onto the raw bagasse and its modified phases

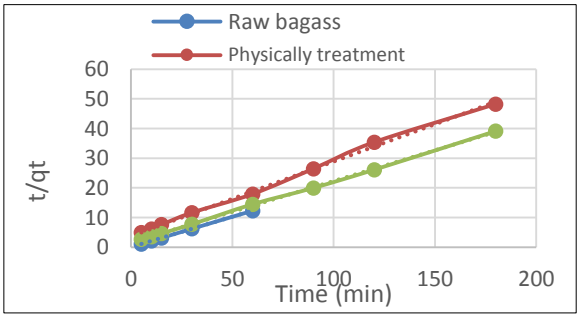

Fig. 12 Pseudo-second-order kinetics plot for the adsorption of methylene blue dye onto the raw bagasse and its modified phases

Table 1: Kinetic parameters for methylene blue adsorption on raw bagasse and its modified phases.

Kinetic Models	Raw bagasse	Physically treatment	Chemically treatment
q_e,exp mg g^{-1}	2.35	2.25	2.325
	Pseudo-First Order		
q_e, mg g^{-1}	1.07229	43.23	4.8
k_1,g/mg.min	0.2037	0.2514	0.2074
R^2	0.9971	0.999	0.9983
	Pseudo-Second Order		
q_e,cal mg g^{-1}	0.909	3.9778	4.8216
k_2, g/mg.min	0.59446	0.0167	0.0272
h, mg g^{-1}.min	0.4908	0.26413	0.6323
R^2	1	0.9978	0.9996

Table 2: Characteristic parameters obtained by Langmuir and Freundlich equations where R^2 is the correlation coefficient.

Adsorbent type	Langmuir Isotherm				Freundlich Isotherm		
	K_L(L/mg)	q_{max}(mg/g)	q_{exp}(mg/g)	R^2	n	K_F(mg/g)	R^2
Raw bagass	10.2	2.1	2.35	0.999	11.46	2.33	0.991
Physicaly modified	4.9	1.9447	2.25	1	7.54	2.37	0.9994
Chemicaly modified	20.57	2.219	2.325	1	16.58	2.33	0.9974

3.6. Isotherm study

Adsorption systems could be investigated using different adsorption isotherms to describe the equilibrium data. Langmuir and Freundlich isotherm models were analyzed in this study. Langmuir isotherm model is compatible when the adsorbent surface is homogenous, and the adsorption occurs on the monolayer. However, the Freundlich isotherm model used when adsorption occupied in a multilayer of the heterogeneous adsorbent surface. The modified phase of equilibrium data, which are applied to Langmuir isotherm Eq.6 and Freundlich isotherms Eq.7, are presented in Fig. (13-14).

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{k_L q_m} \qquad \text{Eq. 6}$$

$$\ln q_e = \ln k_F + \frac{1}{n} \ln C_e \qquad \text{Eq. 7}$$

Where: q_m: the saturation monolayer adsorption quantity (mg. g^{-1}). K_L: the Langmuir adsorption constant (L. mg $^{-1}$). K_F: the Freundlich constant .1/n: the heterogeneity factor.

The values of the adsorption constants, K_L, q_m, q_{exp}, n, and K_F are given in Table 2. Both models agree with experimental data well. Comparing the adsorption capacity of the three adsorbents, the saturation adsorption quantity monotonically gets the highest value when the raw bagasse is treated chemically (2.219 mg.g-1), indicating that the phosphoric group played an essential role in the adsorption process.

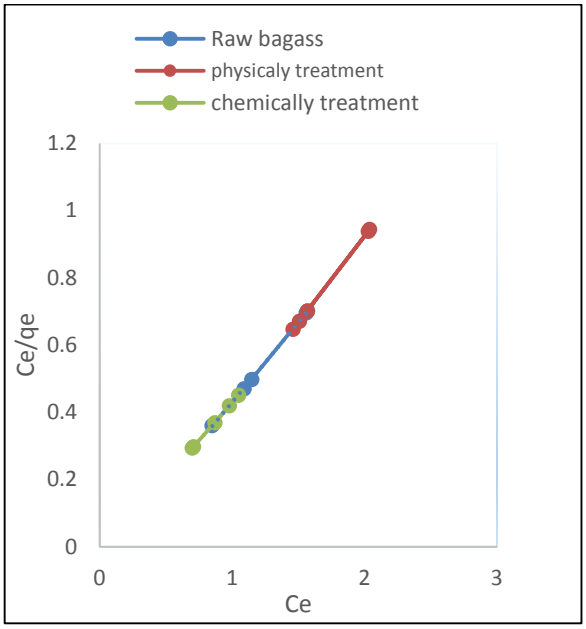

Fig. 13 Langmuir model for adsorption of MB dye on raw bagasse and its modified phases (15 ppm)

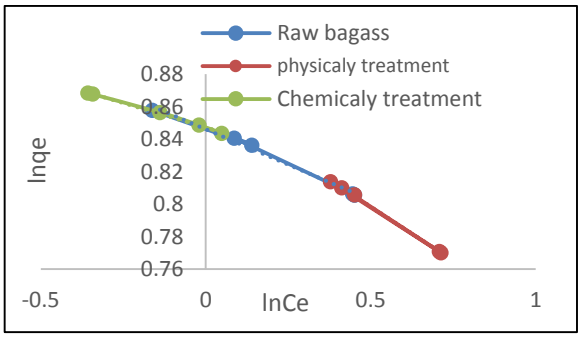

Fig. 14 Frundlish model for adsorption of MB dye on raw 27 bagasse and its modified phases (15 ppm)

4. Conclusion

The modified sugarcane bagasse is a promising adsorber and potentially being used for wastewater treatment. In the present study, the adsorption of MB on raw bagasse and its activated phases were tested. The results show that adsorption slightly decreases with an increase in temperature. The optimum obtained operation conditions using raw bagasse for the maximum percent dye removal (%97.96) were 15 ppm, 100 rpm, 0.3 g/50 ml, and 298 k. The adsorption rate shows the adsorption reaction obeys to the pseudo-second-order kinetic model (average R^2 value 0.999) for the three types of sorbent tested. At the same time, the isothermal study concluded that the results fit both Langmuir and Freundlich models.

Further experiments are in progress to increase the amount of dye removal (q_e) by sugarcane bagasse. Other parameters such as pH variation and other activation additives to the raw material will be studied and reported in detail in a separate paper.

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Conflicts of interest

Not applicable

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الملخص العربي

يعتبر الآن البحث عن المواد المازة للتطبيقات الصناعية والبيئية هي أحد الأهداف قيد التحقيق النشط الآن. يمكن الحصول على الكربون المنشط من المادة العضوية بعد معالجتها كيميائياً أو فيزيائياً. في مصر ، يمكن اعتبار تفل قصب السكرهو أحد المخلفات الزراعية التي تنتج بشكل متكرروفير. في هذه الدراسة ، تم تحضير مادة الامتصاص منخفضة التكلفة من تفل قصب السكر الخام. تم عمل التجربة على دفعات لتحديد تأثير استخدام المخلفات الزراعية (قصب السكر) دون أي تعديل وتفعيلها كيميائيا وفيزيائيا و استخدامها لإزالة صبغة أزرق الميثيلين من محلولها المحضر معمليا. تمت دراسة تأثير كمية المادة المازة ومعدل التقليب ودرجة الحرارة على إزالة الصبغة. أظهرت النتيجة أن نسبة إزالة الصبغة وصلت إلى 99.6٪ عندما كان تركيز الصبغة الإبتدائي 15 جزء في المليون ، وكانت كمية المادة الماصة (المنشط كيميائيًا) 0.3 جم عند درجة حرارة الغرفة. تم دراسة حركيات الامتزاز والايزوثرم لعملية إزالة صبغة بالامتزاز.
و عند تطبيق نماذج لانجمير و فريندليش وجد انهم علي توافق مع عملية الامتزاو محل الدراسة حيث ان R2لها قريبة من الوحدة، وكانت السعة القصوى للامتصاص المحسوبة 2.2 مجم / جم عندما كان تركيز الصبغة الأولي 15 جزء في المليون. علاوة على ذلك ، تم تركيب الرتبة الأولى والرتبة الثانية لفحص حركية الامتزاز ، وحسبت الدرجة الثانية الزائفة أعلى معدل امتزاز لتفل قصب السكر الخام (0.59446 جم / مجم دقيقة).