

Chemical Science International Journal

26(3): 1-11, 2019; Article no.CSIJ.39673

ISSN: 2456-706X

(Past name: American Chemical Science Journal, Past ISSN: 2249-0205)

Equilibrium Adsorption Studies of Methylene Blue onto Caesalpinia pulcherrima Husk-based Activated Carbon

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

Article Information

DOI: 10.9734/CSJI/2019/v26i330096

Fditor(s)

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Reviewers:

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 Complete Peer review History: http://www.sdiarticle3.com/review-history/39673

Original Research Article

Received 26 December 2017 Accepted 13 April 2018 Published 03 April 2019

ABSTRACT

Activated carbon was prepared from Caesalpinia pulcherrima husk (CPH) by chemical activation method using phosphoric acid as an activating agent. The activated sample was characterized based on the physical properties. The experimental data were fitted to Langmuir, Freundlich and Temkin models. The coefficient of determination for Langmuir model $R^2 = 0.967$ was higher compared to Freundlich and Temkin showing monolayer adsorption, and also established that the adsorption of methylene onto CPH based activated carbon can be best described by the Langmuir isotherm model. The energy of adsorption (806.2 kJ/mol) obtained from this model shows clearly that the process is chemisorption. It was observed that the adsorption characteristic indicates a rapid uptake of the adsorbate; the optimum contact time for the adsorption of Methylene blue onto the prepared carbon was 60 minutes. This evidence also points to chemisorption process. These results demonstrate that the carbon derived from CPH can be used as an efficient adsorbent for the adsorption of Methylene blue.

Keywords: Activated carbon; Caesalpinia pulcherrima husk; phosphoric acid; methylene blue; chemisorption.

1. INTRODUCTION

The problems of the ecosystem are increasing with increasing anthropogenic activities. Heavy metal pollution is one of the main problems affecting our ecosystem. Toxic metal compounds do not only contaminate the seas, lakes, ponds and reservoirs but also contaminate underground water in trace amounts by leaching through the soil after rain and snow. Therefore, the earth's waters contain various toxic metals [1]. Heavy metal contamination of water sources is of great concern because of its non-biodegradability and toxic effect to humans, other animals and plants even at very low concentrations [2]. Various chemical and physical analytical methods have been used to remove metal ions. These methods include chemical precipitation, solvent extraction, ion exchange, evaporation, reverse osmosis, electrolysis and adsorption. Among these methods. chemical precipitation, solvent extraction, ion exchange and adsorption were more commonly used [3].

Adsorption using activated carbon has been considered as, possibly, the most cost-effective method for heavy metal ion removal, especially at medium to low concentrations, because the process is simple and chemical consumption or waste generation is not a significant issue [4,5].

Activated carbons form a large and important class of porous solids, which have found a wide range of technological applications [6]. The characteristics and structural properties of activated carbon depend on the physical and chemical properties of the precursor, method of activation, oxidizing agent, time and temperature employed. Activated carbons are carbonaceous material that can be distinguished from elemental carbon by the oxidation of the carbon atoms found in the inner and outer layers [7]. These materials are characterized by their large surface area, the porous surface containing functional group. For this reason, activated carbons are widely used as adsorbents for the removal of organic chemicals and pollutant from air, gases, portable water and also in wastewater treatment [8]. The surface oxygen functional groups can be easily introduced to the carbon by different activation methods including dry and wet oxidizing agents. Wet oxidation methods involve the reaction between the carbon surface and

solutions of oxidizing agents such as phosphoric acid.

(H₃PO₄), Nitric acid (HNO₃), Zinc chloride (ZnCl₂), Potassium hydroxide (KOH), Potassium permanganate (KMnO₄), Calcium chloride (CaCl₂) etc. One of the fastest growing areas in the environmental application of activated carbon to treat effluent from the textile industry. This effluent when discharged defaces the look of natural water and highly toxic [9], also causes harm to mammalian cells by causing kidney tumours [10,11]. Many treatment methods have been employed to remove dyes from wastewater; using physical and chemical methods [12] which are considered expensive in terms of energy and reagents consumption [13,14].

In recent years, the interest in the production of activated carbon from agricultural waste cannot be over emphasized; an added value to waste. Several substances have been screened as alternative to conventional raw materials for the production of activated carbon which include: palm seed coat [15], coconut shells [13], sepiolite pellet [14], and bamboo stem wastes [15] tridax procumbens [16], olive stones [17]. cow bone [18], land snail shell [19], orange waste [20], Terminalia arjuna nuts [21], euphorbia antiquorum L [22], periwinkle shell [23]. Agriculture wastes such as coconut shells, banana peels, oil palm nut shells, rice husks, olive waste cakes, and guava seeds have been reportedly used [10]. The Caesalpinia pulcherrima husk is common in the Nigerian environment and it grows to about 15 m in height and has dark, evergreen, pinnate leaves and produce husks in which lies a number of seeds. husks litter our communities corresponding negative environmental impact. This prompted the need to consider it in producing an activated carbon for the adsorption of a model solution of standard dye (Methylene blue).

2. MATERIALS AND METHOD

2.1 Materials

The carbonaceous precursor used for the preparation of activated carbon (AC) is Caesalpinia pulcherrima husk (CPH) and was collected from the premises of S.S Simon and Jude Seminary, Kuje, Abuja. Prior to use, samples were washed gently with water to

remove mud and other impurities present on the surface and then sun-dried for one week. All chemical reagents used in this work were procured from Steve Moore chemicals, Zaria, Kaduna state. Other materials include a Phosphoric acid H₃PO₄ laboratory standard reagent, distilled water and Equipment include: weighing balance, (Denver Instrument Vecstar Muffle Furnace, Oven (Thermostatic Drying Oven DHG-9202), desiccators, pH meter, UV spectrophotometer and Moisture analyzer).

2.2 Preparation of Activated Carbons

The adsorbents were prepared as described by [24]. Husks from CPH collected after discarding the fruit pulps, were sun-dried, crushed and grinded in a ball mill individually. The grinded samples were sieved to obtain the particles of uniform size, 1.0 mm. The ratio of acid to precursor was 2:1 i.e 200 mL of acid w/v for every 100g of the sample. The preparation of the activated carbon using phosphoric acid was carried out by using predefined conditions of Carbonization temperature, concentration, and particle size and activation time suggested by the software (DOE). The precursor obtained were washed to remove surface bounded impurities and mud then dried. The adsorbents prepared were denoted CPH-PA throughout the work, where PA is Phosphoric acid.

2.3 Characterization of the Adsorbents

2.3.1 Proximate analysis

2.3.1.1 Moisture content

The moisture content was found by oven-drying test method [25]. A sample of carbon was put into a dry, closed capsule (of known weight) and weighed accurately. The capsule is opened and placed with the lid in a preheated oven (145 – 155°C). The sample was dried to constant weight then removed from the oven and with the capsule closed, cooled to room temperature. The closed capsule was weighed again accurately. The percentage difference of weight is expressed as the moisture content of the sample.

2.3.1.2 Percentage volatile matter

The percentage of volatile matter of the AC samples was determined by the standard method [26]. Approximately 1.0 g of the sample was taken in a crucible with cover (of known weight). The covered crucible was placed in muffle

furnace regulated at 950°C for 7 min. Then the covered crucible was cooled to room temperature in a desiccator and weighed. The percentage weight loss was regarded as the percentage of volatile matter.

2.3.1.3 Ash content

To determine the ash content, dried sample of activated carbon was weighed to the nearest 0.1mg and taken into the crucible (of known weight). The crucible was placed in the muffle furnace at 650°C and ashing was considered to be completed when constant weight was achieved. The crucible was cooled to room temperature in a desiccator and the percentage weight of the sample remained was considered as the ash content (ASTM D2866 – 94).

2.3.1.4 Fixed carbon

Fixed carbon is a calculated value and it is the resultant of summation of percentage moisture, ash, and volatile matter subtracted from 100.

Fixed carbon (%) =
$$100 - (moisture, \% + ash, \% + volatile matter, \%)$$
 (1)

2.3.2 Ultimate analysis

The ultimate analysis or elemental analysis was carried out by using CHNS analyzer (Elementar Vario EL CUBE). The percentage of oxygen was calculated by the difference as follows.

Oxygen (%) =
$$100 - (C, \% + H, \% + N, \% + S, \%)$$
 (2)

2.3.3 Activated carbon yield

The yield of activated carbon (AC) was calculated on a chemical-free basis and can be regarded as an indicator of the process efficiency for the chemical activation process. The yield of AC was calculated as the percentage weight of the resultant activated carbon divided by weight of dried precursor.

2.3.4 Bulk density

The bulk density was estimated by placing the product into a graduated cylinder and compacted by tapping on the bench top until an expected volume (cm³) was occupied by mass (g). The

cylinder was tapped on the bench top until the volume of the sample stop decreasing. The mass and volume were recorded and density calculated as equation below [27]

$$\rho = \frac{mass}{volume\ occupied} \tag{4}$$

2.3.5 pH measurement

The 1% solution of the sample was made using deionized water. The pH of the supernatant was obtained after 1 h using a pre-calibrated pH meter (Oaklon pH meter, Model 1100).

2.3.6 Determination of porosity based on swellings procedure

The 0.5 g sorbent was dispersed in 20 mL water (V_1) in a graduated tube with the aid of a shaker. This was further centrifuged for 10 min at 2000 rpm using centrifuge (Baird and Tatlock Auto Bench). The resulting volume was read at V_2 and recorded. The equation 5 was used to calculate the porosity [28]:

Porosity =
$$\frac{V_1}{V_2}$$
 (5)

2.3.7 Determination of titrable surface functional groups

Oxygenated surface functional groups of the prepared activated carbon were determined by Boehm's titration method.

2.4 Adsorption Isotherm Studies

A stock solution of MB was prepared by dissolving 0.5 g in 1000 cm³ of distilled water. Serial dilutions were made to obtain the required lower concentrations. The concentration of MB in the aqueous solution was determined at λ_{max} of 660 nm, using UV-visible spectrophotometer (Helios γ). Methylene blue solution (25 cm³) of known initial concentration and a 0.6 g (particle size of 1.0 mm) of activated carbon were taken in a 150 cm³ Erlenmeyer flasks with airtight stopper. This mixture was agitated in a temperature controlled shaker water bath, at a constant shaking speed for each experiment. The percentage of Methylene blue adsorbed and the amount adsorbed were calculated as [29]:

$$\% Adsorbed = \frac{c_o - c_e}{c_o} X 100$$
 (6)

Where C_o is the initial concentration of the adsorbate (ppm) and C_e is the equilibrium concentration (ppm).

Amount adsorbed
$$(q_e) = \frac{(C_o - C_e)V}{M}$$
 (7)

Where m (g) is the weight of activated carbon used for the adsorption studies and V (cm³) is the volume of the adsorbate. A similar procedure was followed for another set of Erlenmeyer flask containing the same adsorbate concentration without activated carbon was used as blank.

2.5 Adsorption Isotherms

Isotherms give an equilibrium relationship between the amounts of adsorbates adsorbed on the adsorbent surface and its concentration in the solution at a constant temperature. Numerous adsorption models are available in the literature to fit the experimental adsorption data. In this study, the data were fitted using Langmuir and Freundlich models and Temkin models. Each of these models makes uses of a parameter q_e (i.e. adsorption capacity per unit mass of the adsorbent at equilibrium) in mg/g

$$q_{e=\frac{(C_{o-C_e})V}{V}} \tag{8}$$

 C_o = Initial concentration of solution in mg/l C_e = Equilibrium concentration of solution in mg/l V = Volume of solution cm³ X = Amount of adsorbent used in g

2.5.1 Langmuir Isotherm model

The linear form of Langmuir expression:

$$\frac{c_e}{q_e} = \frac{c_e}{Q_o} + \frac{1}{bQ_o} \tag{9}$$

Where C_e is the equilibrium concentration of dye solution (mg/l), q_e is the equilibrium capacity of dye on the adsorbents (mg/g), Q_o is the monolayer adsorption capacity of the adsorbent (mg/g), and b is the Langmuir adsorption constant (L/mg) and is related to the free energy of adsorption. Langmuir model was fitted to the experiment data as shown in the figure below. The activated carbons were taken as 0.6 g for different concentration of Methylene blue. The solution inside the conical flasks was maintained around pH 5 and it was kept in shaker for 60 min.

2.5.2 Freundlich Isotherm model

The Freundlich model is based on the assumption that the Methylene blue is adsorbed on a heterogeneous surface and is not restricted to formation of monomolecular layer.

The linear form can be written as:

$$\ln q_{e=\ln K_f + \left(\frac{1}{n}\right) \ln C_e} \tag{10}$$

Where, K_f and n (dimensionless constants) are the Freundlich adsorption isotherm constants, which indicate the capacity and intensity of the adsorption, respectively.

2.5.3 Temkin isotherm

Temkin isotherm assumes that heat of adsorption decrease linearly with the adsorption onto the surface at a particular temperature and the adsorption is characterized by a uniform distribution of binding energies. Temkin isotherm is expressed in linear form by the following equation [29]:

$$q_e = B_T \ln A_T + B \ln C_e \tag{11}$$

where B_T is related to the heat of adsorption, T (K) is the absolute temperature, R is the universal gas constant (8.3143 J/mol), b indicates the adsorption potential of the adsorbent (J/mol), A_T is the equilibrium binding constant (L/mg). The parameters for the Temkin model are obtained from the plot of q_e versus In $C_{\rm e}.$

3. RESULTS AND DISCUSSIONS

Activated carbons were prepared from CPH as a precursor with chemical activation using Phosphoric acid as the activating agent. Phosphoric acid is a common activating agent in activated carbon manufacturing industries. The selection of this chemical in this study was informed by the fact that it is a cleaner production technology in compared to the use of ZnCl₂, KOH and other chemical agent frequently being used. The activated carbon was characterized by standard methods. Scanning Electron Microscopy (SEM) was used to study the morphological structure of the prepared activated carbons.

3.1 Physicochemical Analysis of CPH

The results obtained from the physicochemical parameters of the CPH are presented in Table 1 The percent dry matter and moisture of CPH are 83.76 % and 4.34 %, respectively. The percent moisture was lower than 6.48% reported by Erhan et al. [30] for cornelian cherry and lower than 5.75% reported by Shakirullah et al. [31] for

sawdust. This high value may be as a result of its hydrophilic nature.

The pH of CPH was found to be 5.32. This value falls within the acceptable pH conditions for adsorption [32]. Bulk density determines to a large extent the length of the filtration cycle of activated carbon [33]. The bulk density for CPH was found to be 0.48 g/cm³. This value according to Erhan et al. [30] suggests that CPH is a good adsorbent in terms of volume activity. The ash content and porosity value of the sample were found to be within the range reported by other researchers [34]. This is favourable because it is relatively low. The lower the ash content (2.43) of CPH suggests that it is a better material for adsorption since the ash serves as interferences during the adsorption process [34].

Table 1. Physicochemical parameters of CPH-PA

Parameters	Values
pH of 1% solution	5.32
Bulk density (g/cm ³)	0.48
Moisture content (%)	4.34
Dry matter (%)	83.76
Volatile matter (%)	10.62
Ash content (%)	2.43
Porosity	0.87
lodine adsorption number (mM/g)	0.17

Table 2. Ultimate analysis of CPH-PA

Parameters	Values
С	88.69
Н	1.17
N	1.85
S	1.58
others	6.71

The iodine adsorption number (IAN) measures the adsorption of iodine from an aqueous solution. It is a measure of microspores and it is used as an indication of the total surface area. Adsorbents with high iodine number perform better in removing small sized contaminants. It is the most fundamental parameter used to characterize the performance of activated carbon. High value indicates high degree of activation [27]. From the result presented in Table 1 above, the IAN for MLH estimated in mM iodine per gram of adsorbent is fairly high (0.17), as compared to that reported by Itodo [35] for Shea nut shells (0.1338 - 0.1505) and groundnut shells (0.1115 - 0.1394). It thus

implies that MLH presents high degree of activation and high affinity for small sized contaminants.

3.2 Ultimate Analysis of Activated Carbon

The ultimate analysis gives the composition of the biomass as well as the prepared activated carbon in wt% of carbon, hydrogen and nitrogen (the major components) and others (sulfur and oxygen). The results obtained for ultimate analysis in this study are listed in Table 1 above.

All the samples have moderate nitrogen content. It is observed that the carbon content increased significantly after activation of the raw precursors revealing that phosphoric acid activation method used here is suitable to develop activated carbon from the selected precursors which can subsequently enhance the uptake capacity for the metals from contaminated water. From the ultimate analysis, it is observed that the elemental carbon content is slightly higher than fixed carbon determined by proximate analysis. With the increase of temperature impregnation ratio, hydrogen content is decreasing. It was observed that the hydrogen content of phosphoric acid activated bamboobased activated carbon was decreasing with the increase of temperature and impregnation ratio [36].

3.3 Determination of Titrable Surface Functional Groups

The adsorption capacity of activated carbon depends upon the chemical nature of surface functional groups which are present in the form of acidic and/or basic oxides. The acidic character of a functional group depends on its chemical environment i.e. the size and shape of the poly-aromatic layers, the presence and position of other constituents and the charge of neighbouring dissociated groups. The presence of carboxylic groups, lactonic groups, hydroxyl groups of phenolic character etc, on an adsorbent surface makes it acidic in nature. Different acidic functional groups differ in their acid strength and can be easily differentiated from one another by titration with the aqueous solutions of selected bases NaOH, Na₂CO₃, NaHCO₃ and C₂H₅ONa according to Boehm's method. Sodium hydroxide (NaOH) is a strong base in aqueous medium that can neutralize acids of strong or weak ionization potentials. The NaOH titration can be used to measure the surface acidic character of activated carbon

samples due to the presence of carboxylic acids, phenols, lactones and other groups with acidic or ionizable hydrogen atoms. The surface functional groups of the prepared carbon samples are shown in Table 3 below which shows that the dominant surface functional groups were acidic in nature. The more acidic surfaces of ACs could be due to the presence of an increased oxygen-containing acidic groups primarily quinones, lactones, phenols and carboxylic functional groups. These groups might have been introduced during phosphoric acid activation [37].

Table 3. Titrable surface functional groups for CPH-PA

Properties	CPH-PA
Phenolics + Lactones x 10 ⁻³	1.56
(m.mol.g ⁻¹)	
Carboxylics x 10 ⁻³ (m.mol.g ⁻¹) Quinone x 10 ⁻³ (m.mol.g ⁻¹)	1.83
Quinone x 10 ⁻³ (m.mol.g ⁻¹)	4.14
Basic groups x 10 ⁻³ (m.mol.g ⁻¹)	0.87

The surface structures of the activated carbons have burnt out pores with a tunnel or honeycomb-like structures as shown in Fig. 1b. The sample is characterized by a smooth surface with many orderly pores developed. This is as a result of lack of tars and other impurities that could clog up the pores and inhibit the development of pore structures. The BET surface area as obtained from nitrogen gas adsorption was 1069 g/m². The nitrogen gas adsorption also showed that the adsorption was characteristic of type 1 of IUPAC isotherm classification. This is indicative of predominantly porous adsorbent. The micropore volume and the total pore volume were 0.438 and 0.5789 cm³g⁻¹ respectively.

3.4 Effect of Contact Time and Initial Concentration on Adsorption of MB CPH-PA

The effect of adsorption on contact time was studied using fixed amount (0.6 g) of adsorbents on 20 mg/l Methylene blue solution in a fixed volume (100 ml). It was observed that adsorption increases with increase in contact time. Initially, within 10 min exponential, increase in adsorption was reflected till maximum slope reached in next 10 min. Thereafter a steady state equilibrium was obtained after a very long time.

It is clear that the percent adsorbed increased with contact time but however decreased with

increase in initial concentration. The adsorption characteristic indicates a rapid uptake of the adsorbate as shown by the curves. adsorption rate however increased The marginally after the first 20 minutes to a nearly constant value with increase in contact time. This agrees with the report of other investigators in the literature [38,39,40]. The percent adsorbed was maximum at 60 minutes at most initial concentration, hence 60 minutes is the optimum contact time for the adsorption of methylene blue onto the prepared carbon. The percent adsorbed decreased from 94.80 to 91.11 percent after the first twenty minutes of contact of the MB with the adsorbent as the initial concentration increased from 20 to 40 mg/l.

3.5 Adsorption Isotherms

Two important physicochemical aspects for the evaluation of adsorption process as a unit operation are the equilibria of the adsorption and the kinetics. The adsorption equilibrium usually presented as adsorption isotherm is useful for predicting the performances of activated carbon. According to [41], adsorption capacity is a prime factor to be considered in the optimization of activated carbon. The equilibrium relationships between adsorbate and adsorbent are described by adsorption isotherms, usually, the ratio between the quantity adsorbed and that remaining in solution at a fixed temperature at equilibrium [42]. The analysis of isotherm data by

fitting them into different isotherm models is an important step to find the suitable model that can be used to reproduce the experimental result obtained and hence for design purposes [43].

Table 4. Adsorption isotherm constants parameters

Langmuir	Values
Q _o	10.98
В	5.05
R_L R^2	0.0052
R^2	0.967
Freundlich	
K_f	0.263
1/n	1.102
R^2	0.522
Temkin	
B _T (Kj/mol)	806.2
$K_{T}(dm^{3}/g)$ R^{2}	0.305
R^2	0.535

The Langmuir isotherm constants, separation factor and the correlation coefficient are shown in Table 4 below. The value of adsorption capacity, Q_{o} , and the constant relating to the rate of adsorption, b, as obtained from the slope and intercept of the plot were 10.98 mg/g and 5.05 L/mg, respectively (Table 4). A correlation coefficient, as high as 0.967 was observed, indicating that the experimental data fit for the Langmuir model.

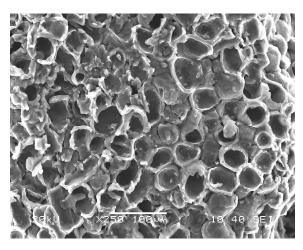
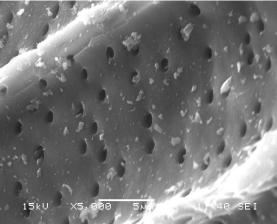


Fig. 1. (a) MLH Precursor



(b) MLH-PA AC

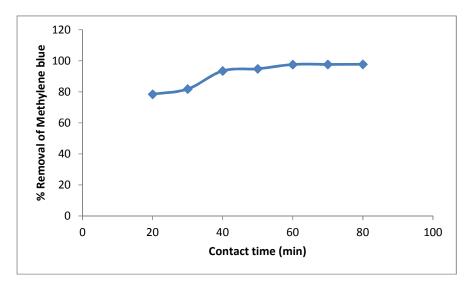


Fig. 2. Study of removal of Methylene blue using CPH-PA

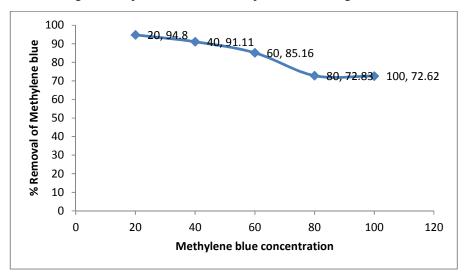


Fig. 3. Percentage removal of Methylene blue vs. different concentration for CPH-PA

An important characteristic of the Langmuir model is a dimensionless parameter called the separation factor, R_L and expressed as (Hameed et al. 2006):

$$R_L = \frac{1}{1 + bC_O} \tag{12}$$

Where b is the Langmuir constant expressing the adsorption rate and C_{o} is the highest initial concentration. The R_{L} parameter is considered as a reliable indicator of adsorption characteristic. It indicates the shape of the isotherm and the nature of the adsorption process as given below [40].

 R_L Value Nature of the process $R_L > 1$ Unfavourable

 $R_L = 1$ Linear 0 < RL < 1 Favourable RL = 0 Irreversible

The $R_{\rm L}$ value was 0.0052. The $R_{\rm L}$ value for the studied system was found to be between 0 and 1which is indicative of favourable adsorption of Methylene blue onto the adsorbent.

The Temkin isotherm constant for the adsorption of MB from aqueous solution onto CPH shell based activated carbon is shown in Table 4. The low correlation coefficient (0.52) was characterized by the fact that the plots don't fit into the model. The low value indicates a weak interaction between the adsorbate and the adsorbent. The Temkin isotherm constant shows

that the value of B_T , is related to the heat of adsorption, was 806.2 kJ/mol. This value is indication of chemisorptions. The Temkin adsorption potential, K_T of the adsorbent was 30.5 (dm³/g).

The Freundlich isotherm parameters, K_f and 1/n which measure the capacity of the adsorbent and adsorption intensity/ surface heterogeneity are obtained from the intercept and the slope of the plot, respectively, and are shown in Table 4. The K_f value was 0.262 while the value of 1/n was 1.102. Generally, 1<1/n<10 is characteristic of cooperative adsorption and indicative of favourable adsorption process as reported in the literature [44,45]. In cooperative adsorption, the formation of the second layer is ruled out as there is no obvious phase change [46]. Freundlich model didn't yield a good fit with a correlation coefficient value of 0.535.

4. CONCLUSION

This investigation has shown that Caesalpinia pulcherrima husk could be a good precursor for preparation of a low cost activated carbon for removal of dye from wastewater. This is shown by the results of the physicochemical properties analysis of the prepared activated carbon. The pH and ash content is low; the BET surface area is adequate and SEM analysis shows the development of adequate pores during the activation process. The carbon is, therefore, suitable for the adsorption of Methylene blue from the aqueous phase. It was also observed that the surface of the prepared activated carbon is heterogeneous with a variety of functional groups present; as such the adsorbent could be effective for adsorption of a variety of impurities from the solution. The present study established that the adsorption of Methylene Caesalpinia pulcherrima husk based activated carbon could be best described by the Langmuir isotherm model with a characteristic correlation coefficient of 0.967. The energy of adsorption obtained from this model shows clearly that the process is chemisorptions. This is attributed by the fact that the nitrogen gas adsorption also showed that the adsorption was characterize by type 1 of IUPAC isotherm classification and indicative of a predominantly porous adsorbent. It is observed that the adsorption characteristic indicates a rapid uptake of the adsorbate, the optimum contact time for the adsorption of Methylene blue onto the prepared carbon is 60 minutes. This evidence also point chemisorptions process. Activated carbon

prepared from *Caesalpinia pulcherrima* husk was recorded to be a promising adsorbent for the removal of Methylene blue and this research will serve as a basic data for further research on the use of above-mentioned adsorbent for adsorption studies.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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