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$FeCl_3$-Activated Carbon Developed from Coconut Leaves: Characterization and Application for Methylene Blue Removal

(Karbon Teraktif-$FeCl_3$ daripada Daun Kelapa: Pencirian dan Aplikasi terhadap Penyingkiran Metilena Biru)

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ABSTRACT

In this study, coconut leaves were used as a starting material for the production of activated carbon by thermal carbonization using $FeCl_3$-activation method. The characterization of coconut leaves-$FeCl_3$ activated carbon (FAC) were evaluated by bulk density, ash content, moisture content, point-of-zero charge (pH_{pzc}) analysis, iodine test, scanning electron microscopy (SEM), Fourier transform infrared (FTIR) and elemental (CHNS-O) analysis. The effect of the adsorbent dosage (0.02-0.25 g), initial pH (3-11), initial dye concentrations (30-350 mg/L) and contact time (1-180 min) on the adsorption of the methylene blue (MB) at 303 K was performed via batch experiments. The Pseudo-Second Order (PSO) describes the kinetic model well whereas the Langmuir isotherm proved that adsorption behavior at equilibrium with maximum adsorption capacity (q_{max}) of 66.00 mg/g.

Keywords: Activated carbon; adsorption; coconut leaves; ferric chloride; methylene blue; thermal activation

ABSTRAK

Dalam kajian ini, daun kelapa telah dipilih sebagai bahan pemula bagi penghasilan karbon teraktif melalui penkarbonan haba dengan menggunakan $FeCl_3$ sebagai agen pengaktif. Pencirian karbon teraktif (FAC) dianalisis dengan menggunakan mikroskop elektron imbasan (SEM), transformasi Fourier inframerah (FTIR) dan analisis elemen (CHNS-O). Beberapa parameter yang mempengaruhi penyahwarnaan metilena biru pada suhu 303 K seperti dos bahan penjerap (0.02-0.25 g), pH awal (3-11), kepekatan (30-350 mg/L) dan masa (0-180 min) telah dikaji dan dioptimumkan dengan menggunakan kaedah uji kaji berkumpulan. Model kinetik Pseudo-Pertama dan Pseudo-Kedua telah digunakan untuk menganalisis mekanisme yang terlibat dalam proses penjerapan. Nilai korelasi (R^2) yang ditunjukkan oleh FAC terhadap model kinetik tertib Pseudo- Kedua lebih tinggi berbanding tertib Pseudo-Pertama. Data isoterma penjerapan dikaji dengan model isoterma seperti Langmuir, Freundlich dan Temkin. Proses penjerapan dikenal pasti mengikuti model Langmuir dengan kapasiti penjerapan maksimum (q_{max}) dicapai sebanyak 66.00 mg/g.

Kata kunci: Daun kelapa; ferik klorida; karbon teraktif; metilena biru; pengaktifan haba; penjerapan

INTRODUCTION

Wastewaters from textile, rubber, paper, plastics, leather and food industries contain dyes used to color their final products. The discharge of dye-contained wastewaters into ecosystem is a dramatic source of aesthetic pollution, eutrophication and perturbation in aquatic life as most of dyes are highly visible, stable and unaffected to chemical, photochemical as well as biological degradation (Jawad et al. 2017a, 2016a; Mubarak et al. 2017). Methylene blue (MB) is a basic dye with favorable water solubility and the most commonly used for dyeing of textiles and leather, printing calico, printing cotton and biological staining methods (Jawad et al. 2016b). MB has various harmful effects on human beings, such as eye irritation, gastrointestinal irritation and nausea upon ingestion, including vomiting and diarrhea (Jawad et al. 2017b), so it is of utmost importance to be removed from wastewaters. Various conventional technologies have been tested for the removal of dyes from industrial effluents and wastewaters, including bioremediation (Khataee et al. 2012), electrochemical degradation (Fan et al. 2008), cation exchange membranes (Wu et al. 2008), Fenton chemical oxidation (Woo et al. 2013) and photocatalysis (Jawad et al. 2015, 2016c). Most of these methods, nevertheless, pose techno-economical limitations for field-scale applications (Akbal 2005). Comparatively, adsorption has been proved to be a well- established and most widely used technique among other water purification processes. Adsorption-based treatment with appropriate adsorbent materials shows high performance and selectivity, flexibility and simplicity of design, convenience of operation without producing harmful by-products as well as economically cost effective (Jawad et al. 2017c).

Activated carbons (AC) are materials containing large surface area, well-developed porosity and various functional groups. Therefore, AC has been widely utilized in versatile applications such as gas separation, solvents recovery, gas storage, super capacitors electrodes, catalyst support and adsorbent for organic and inorganic pollutants from drinking water (Xu et al. 2014). However, a high

cost of AC production limits its application in various technologies. Recognizing this economic obstacle, many investigators have been made extensive efforts in low-cost alternatives to activated carbon from a range of carbonaceous precursors, such as lignocellulosic materials (Jawad et al. 2016d), biopolymer (Marrakchi et al. 2017), coal (Gao et al. 2016), char (Acosta et al. 2016) and fruit peels (Jawad et al. 2017d). The textural properties and adsorption capacities of carbonaceous materials are mainly depend on the nature of the starting martial, activation method, type of activator and preparation conditions (Liu et al. 2016).

Coconut is a versatile plant species. Other than food source, coconut has been used as fuel wood, drink, edible oil, fiber, animal feed and building materials. Some coconut by-products, such as husks (Johari et al. 2016), shells (Cazetta et al. 2011; Isah et al. 2015), mesocarp (Vieira et al. 2010, 2009), frond (Njoku et al. 2014) and coir (Etim et al. 2016) have been used as the potential precursors for the production of activated carbons. Its leaves however have limited function. Normally, the leaves are cultivated for handicraft and decorative purpose only. Although the literature for superiority of coconut leaves structure is not much elaborated, but in general, agriculture waste components such as hemicellulose, lignin, lipid, protein, simple sugar, water, hydrocarbon and starch contain functional groups with a potential sorption capability (Bhatnagar & Sillanpää 2010; Bhatnagar et al. 2015). In particular, the goal of this study was to prepare activated carbon from coconut leaves by thermal carbonization using $FeCl_3$-activation. The feasibility of the FAC was tested for MB adsorption.

Materials and Methods

MATERIALS

The coconut leaves used for the preparation of activated carbon were collected from Universiti Teknologi MARA (UiTM), Arau Campus, Perlis, Malaysia. Chemicals used in this study were ferric chloride (HmbG), sodium hydroxide pellete (HmbG), hydrochloric acids (QrëC), sodium chloride (HmbG), sodium thiosulphate (HmbG), iodine pearl (HmbG), potassium iodate (HmbG), starch (HmbG) and methylene blue (R&M; molecular weight: 319.85 g/mol; molecular formula: $C_{16}H_{18}N_3SCl$; λ_{max}: 661 nm). All chemicals were used directly without further purification. Ultrapure water of 18.2 MΩ/cm was used throughout this work.

PREPARATION OF COCONUT LEAVES-$FECL_3$ ACTIVATED CARBON (FAC)

The coconut leaves were washed with tap water to remove dust and any impurities before drying in an oven (Memmert, model UFB-400) at 80°C for 48 h. The dried sample was cut into smaller pieces and ground into powdered form. The coconut leaves-$FeCl_3$ activated carbon (FAC) was prepared by mixing $FeCl_3$ with a biomass/$FeCl_3$ impregnation ratio of 1:1 (wt. %) with occasional stirring and then kept in an oven for 24 h at 110°C. Then, the impregnated sample was placed in a stainless steel vertical tubular reactor in furnace. The carbonization process was conducted under high purified nitrogen gas (99.99%) at 700°C under the pressure of 1 atm for 60 min. The product was then cooled to room temperature and washed with 3 mol/L of HCl solution to eliminate ash followed by hot distilled water until the filtrate turn neutral pH. The FAC was then dried in an oven at 110°C for another 24 h. After that, the FAC powder was sieved to obtain a particle size range of 150 μm-212 μm. Finally the FAC was stored in tightly closed bottles for subsequent use. The percentage yield of the activated carbon was calculated by (1):

$$\text{Yield } (\%) = \frac{W_f}{W_o} \times 100 \quad (1)$$

where W_f and W_o are the final weight of activated carbon product (g) and the weight of dried activated carbon (g), respectively.

CHARACTERIZATION OF FAC

The physical properties of coconut leaves-$FeCl_3$ activated carbon (FAC) were evaluated through bulk density, ash content, moisture content, point-of-zero charge (pH_{pzc}) and iodine test. Bulk density, ash content and moisture content were determined according to procedure described by Ahmed and Dhedan (2012) while pH_{pzc} was estimated using a pH meter (Metrohm, Model 827 pH Lab, Switzerland), as described by Lopez-Ramon et al. (1999). Iodine test was determined from ASTM D4607-14. The surface physical morphology of FAC was examined by using Scanning Electron Microscope (Leica Cambridge S360). Fourier transform infrared (FTIR) spectrometer was used to verify the presence of surface functional groups in the 4000 - 500 cm^{-1} wavenumber ranges and the elemental (CHNS-O) analysis was carried out using elemental analyzer Flash 2000 Model.

BATCH ADSORPTION EXPERIMENTS

The batch adsorption experiments of MB onto coconut leaves-$FeCl_3$ activated carbon (FAC) were conducted in a set of 250 mL Erlenmeyer flasks containing 100 mL of MB solution. The flasks were covered and agitated in an isothermal water bath shaker (Memmert, waterbath, model WNB7-45, Germany) at shaking speed of 120 strokes/min at 303 K until equilibrium was achieved. Batch adsorption experiments were carried out by manipulate several experimental variables such as such as adsorbent dosage (0.02 to 0.25 g), initial pH (3 to 11), initial dye concentrations (30 to 350 mg/L) and contact time (0 to 180 min) to determine the best conditions for MB adsorption. After the stirring, the supernatant was collected with 0.2 μm Nylon syringe filter and the concentrations of MB were

analyzed at different time interval using HACH DR 2800 Direct Reading Spectrophotometer at a wavelength of 661 nm. The blank test was carried out in order to account for color leached by the adsorbent and adsorbed by the glass containers, blank runs with only the adsorbent in 100 mL of doubly distilled water and 100 mL of dye solution without any adsorbent were conducted simultaneously at similar conditions. The adsorption capacity at equilibrium, q_e (mg/g) and the percentage of color removal, CR (%) were calculated as stated in (2) and (3), respectively.

$$q_e = \frac{(C_o - C_e)}{W} \quad (2)$$

$$CR\% = \frac{(C_o - C_e)}{C_o} \times 100 \quad (3)$$

where C_o is the initial dye concentration (mg/L); C_e is the dye concentration at equilibrium (mg/L); V is the volume of dye solution used (mL); and W is the dry mass of the adsorbent used (g). Adsorption experiments were conducted in duplicate under identical conditions and the results are reported as average values.

Results and Discussion

PHYSICOCHEMICAL CHARACTERISTIC OF FAC

Table 1 shows the physicochemical properties of coconut leaves-$FeCl_3$ activated carbon (FAC). Bulk density test is conducted to determine the quality of activated carbon. This test related to fiber content presence in the precursor. High fiber content increased bulk density and contribute greater mechanical strength (Balakrishnan & Satyawali 2007). In contrary for the present work, low bulk density (0.36 g/mL) is obtained to indicate low amount of MB the FAC can hold per unit volume.

Ash is mineral additives, which is not chemically combined with the carbon surface. It contains of various mineral constituents and become highly concentrated during activation process. As for activated carbon, the threshold limit of ash content should be less than 15% (De Gisi et al. 2016). Activated carbon with low ash content suggests the precursor have low extractives with little or no wax and resin (Johari et al. 2016). Meanwhile, activated carbon with high ash content has weaker mechanical strength and has the tendency to effects adsorptive capacity (Abdullah et al. 2001). The result in Table 1 indicates that ash content of FAC is low (0.16%), suggesting that the raw material used is a good source for activated carbon production.

Porous materials have the ability to absorb moisture (Baseri et al. 2012). Although, adsorptive power of activated carbon is not influenced by its moisture content, excessive moisture can dilutes activated carbon and affect the actual weight required during the adsorption

TABLE 1. Physicochemical characteristics of FAC

Typical Properties	
Bulk density (g/mL)	0.36
Iodine number (mg/g)	653.00
Proximate analysis (wt. %)	
Ash content (%)	0.16
Moisture content (%)	0.59
Percentage yield (%)	35.00
pHpzc	4.2
Ultimate analysis (wt. %)	
C	61.65
H	1.84
N	0.57
S	0.11
O (by difference)	35.83

experiment (Baseri et al. 2012). Hence, moisture content of activated carbon should be maintained as low as possible. In this work, moisture content of FAC is low with only 0.59%.

The surface charge of FAC was evaluated by point-of-zero charge (pH_{pzc}) analysis, where pH_{pzc} for FAC was 4.2, indicative of its acid character. This observation reconfirms the availability of the acid active groups on the FAC surface, according to the FTIR results in Figure 2(a). A positive charge of FAC surface can be obtained at pH environment below the pH_{pzc}, preferring the uptake of negatively charged species. On the other hand, a negative charge of FAC surface can be obtained at pH levels beyond the pH_{pzc}, preferring the adsorption of MB as cationic species.

Iodine test analysis is fundamental technique used to determine surface area approximation and porosity measurement of activated carbon (Somasekhara Rao et al. 2005). The test is an index of the ability of the adsorbent to adsorb small molecules, expressed in milligrams of iodine adsorbed per gram of activated carbon. Activated carbon has iodine number varies in the range from 500.00 to 1200.00 mg/g (Bonomo 2008) and excellent activated carbon is expected to have iodine number equal to or higher than 900.00 mg/g (Benadjemia et al. 2011). FAC indicated a moderately high iodine number with 653.00 mg/g.

The elemental analysis of FAC shows high carbon level (61.65%) while the content of hydrogen (1.84 %), nitrogen (0.57 %), sulphur (0.11%) and oxygen (35.83%) indicate the opposite. The trend was observed due to the partial decomposition of volatiles compounds and degradation of organic substances leaving a high purity carbon synthesized from coconut leaves.

SEM ANALYSIS OF FAC

Scanning Electron Microscopy (SEM) with a magnification of 3.0 KX was used to analyze the surface structure of coconut leaves-$FeCl_3$ activated carbon (FAC) before and after adsorption of MB. In Figure 1(a), it is observed that the availability of pores structure within the carbon. During the

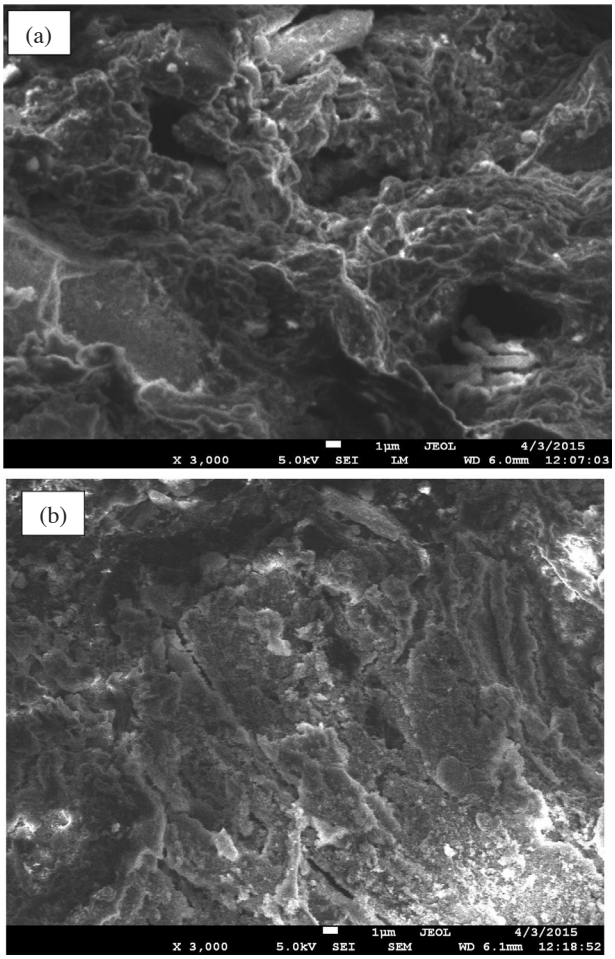

FIGURE 1. SEM Micrograph of (a) FAC and (b) FAC-MB

activation process, chemical activator from the activated carbon undergoes evaporation thus creating framework of pores (Uçar et al. 2009). Before adsorption of MB take place, the surface of the FAC has heterogeneous structure with pore of various sizes. The presence of these cavities allowed MB molecules diffused and clogged into the pore of FAC. This theory was supported by Figure 1(b) where the surface of FAC after adsorption of MB became denser and smoother due to loading of MB on the FAC surface.

FTIR ANALYSIS OF FAC

Fourier transform infrared (FTIR) spectral analysis was performed to determine the functional group of the coconut leaves-$FeCl_3$ activated carbon (FAC). Figure 2(a) shows the IR spectrum of FAC before MB adsorption. IR bands at 1560.00 cm^{-1} assigned to C=C vibration in aromatic ring and 1090.00 cm^{-1} indicate alkane group (Benadjemia et al. 2011). The IR band around 1400.00 cm^{-1} relates to the asymmetric stretching of the sulphonic acid groups. After MB adsorption (Figure 2(b), the spectrum show changes in band intensity in which the functional group is shifted in frequency and form new peak as MB molecules bound onto surface of FAC. The peaks at 1090.00 and 1560.00 cm^{-1} shifted to higher band of 1103.00 and 1570.00 cm^{-1}, respectively. New band appear at 2290.00 cm^{-1} attributed to the aliphatic C-H stretching such in an aromatic methoxyl group, methyl and methylene groups of side chain (Reffas et al. 2010).

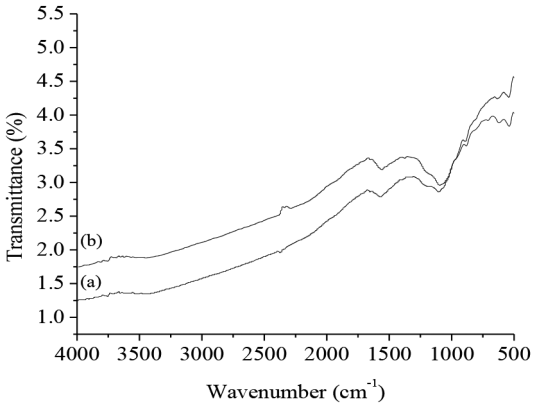

FIGURE 2. FTIR spectra of (a) FAC and (b) FAC-MB

BATCH ADSORPTION STUDY

Adsorption studies emphasis on the effect of parameters that have a great impact on the adsorption process. Some of the parameters include adsorbent dosage, initial pH, initial dyes concentration and contact time.

Effect of Adsorbent Dosage The adsorbent dosage parameter is important to be studied in order determine the capacity of an adsorbent for a given concentration of dye in solution. Figure 3 shows the effect of coconut leaves-$FeCl_3$ activated carbon (FAC) dosage on the removal of MB. Based on the plotted graph, the percentage removal of MB increased as FAC dosage increased. This is due to increased FAC surface area and availability of more adsorption sites resulting from the increase dosage of FAC (Hamdaoui & Chiha 2007). In this study, the highest removal of MB by FAC was recorded at 0.1 g with 74.87% removal. The further increase of FAC dosage beyond 0.1 g did not show any obvious changes.

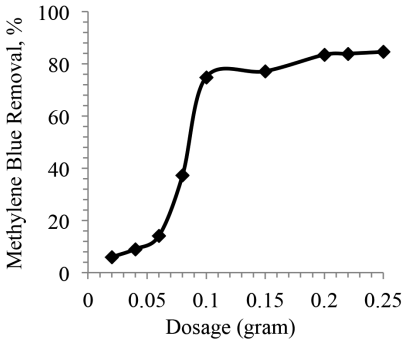

FIGURE 3. Effect of FAC dosage on the MB removal

MB concentration: 100 mg/L; V: 100 mL; pH: 5.6 (Unadjusted); Time: 60 min; Rotational speed: 120 strokes/min; T: ±303 K]

Effect of Initial pH The pH of solution is expected to influence the adsorption capacity of dyes due to its impact on both the surface binding-sites of the adsorbent and the ionization process of the dye molecule (Ncibi et al. 2007). As illustrated in Figure 4, MB uptake by FAC was not affected by pH within the range of 3 to 11 due to buffering effect of the adsorbent. Similar result have been reported for the adsorption of MB by coconut leaves (Jawad et al. 2017b; Rashid et al. 2016), mango peel (Jawad et al. 2017d), *Parthenium hysterophorus* (Lata et al. 2007), *Prosopis cineraria* sawdust (Garg et al. 2004) and *Posidonia oceanica* (L.) fibres (Ncibi et al. 2007). Therefore, the pH value of unadjusted MB solution (pH 5.6) was used throughout this study.

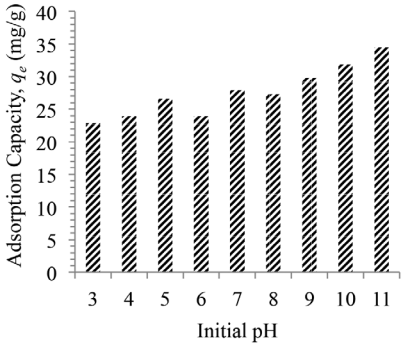

FIGURE 4. Effect of pH on the adsorption capacity of MB by FAC

Adsorbent dosage: 0.1 g; MB concentration: 100 mg/L; V: 100 mL; Time: 60 min; Rotational speed: 120 strokes/min; T: ±303 K

Effect of Concentration and Contact Time The effect of the initial dye concentration depends on the immediate relation between the dye concentration and the available binding sites on the surface of the adsorbent (Etim et al. 2016). In this study, the effect of adsorption capacity was investigated with the initial MB concentration range from 30 to 350 mg/L at temperature of 303 K. Figure 5 shows the results for the adsorption capacities of MB by FAC at various concentrations. The amount of MB adsorbed increased

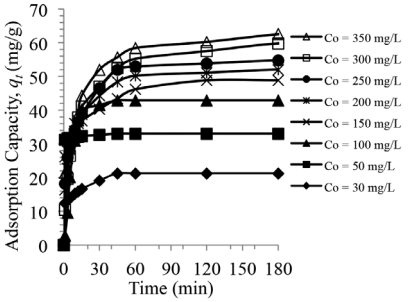

FIGURE 5. Effect of MB concentration on the adsorption capacity of MB by FAC

Adsorbent dosage: 0.1 g; V: 100 mL; pH: 5.6 (Unadjusted); Rotational speed: 120 strokes/min; T: ±303 K

rapidly from 21.29 to 64.38 mg/g with increase of MB concentration from 30 to 350 mg/L. As MB concentrations increase, collision rate between MB cations and FAC also increase. Therefore, more MB cations were transferred to the FAC surface. At higher MB concentration, more time was required to reach equilibrium since there was a tendency for the adsorbate to penetrate deeper within the interior surface of the FAC and occupy more active adsorption sites.

ISOTHERM STUDY OF FAC

The application of adsorption isotherm are useful to predict the interaction between the amount adsorbate adsorbed by the adsorbent (q_e) and the adsorbate concentration remaining in the solution after the system achieved the equilibrium state (C_e) (Njoku et al. 2014). In this study, three isotherm models, namely Freundlich (1906), Langmuir (1918) and Temkin (Temkin & Pyzhev 1940) were employed. The parameters of the isotherm models were listed in Table 2. Based on calculated data, Langmuir model is found to be linear over whole concentrations ($R^2 \geq 0.99$) and assumed that adsorption occurs over a surface with homogenous energy sites, which are equally available for interaction. The monolayer adsorption capacity (q_{max}) for FAC with MB was compared with other types of $FeCl_3$-treated AC adsorbents in Table 3.

TABLE 2. Isotherm parameters for the adsorption of MB by FAC at ±303 K

Langmuir Isotherm	q_{max} (mg/g)	66.00
	k_L (L/mg)	0.04
	R^2	0.99
Freundlich Isotherm	1/n	0.24
	k_F $[(mg/g)\ (L/mg)^{1/n}]$	15.90
	R^2	0.97
Temkin Isotherm	B	9.43
	k_T (L/mg)	2.07
	R^2	0.96

TABLE 3. Comparison of adsorption capacities for MB onto different activated carbons prepared by $FeCl_3$ activation

$FeCl_3$-treated activated carbons	q_{max} (mg/g)	References
Coconut leaves	66.00	This study
Coffee husk	75.00	Oliveira et al. 2009
Iron impregnated AC	32.49	Shah et al. 2014
Iron impregnated AC	30.61	Shah et al. 2015
Acrylic fibrous	20.61	Naeem et al. 2016

KINETIC STUDY OF FAC

In order to investigate the potential rate controlling step, two kinetic models namely Pseudo-First Order (PFO) and Pseudo-Second Order (PSO) were tested on experimental data. Table 4 lists the kinetic parameters for the adsorption of MB by FAC. The conformity between the experimental and the calculated data was determined by the correlation coefficient (R^2). The R^2 values are greater ($R^2 \geq 0.99$) for PSO model and its $q_{e,cal.}$ values are in agreement with the values of $q_{e,exp.}$ Therefore, for the adsorption of MB by FAC, PSO model shows a better fit compare to the PFO model. The analysis of kinetic data by the PSO showed that the rate- controlling step is chemisorption in which involving valence forces through the exchange or sharing of electrons between the adsorbate molecules and the surface functional groups of adsorbent (Ahmed & Dhedan 2012; Gupta 2009).

CONCLUSION

This study investigates the feasibility of coconut leaves as a new and low-cost precursor for the preparation of activated carbon using $FeCl_3$ as chemical activator via thermal method. FAC has an iodine number of 653.00 mg/g. The adsorption experiments showed the adsorption isotherms are well described by the Langmuir model where the maximum adsorption capacity (q_{max}) is 66.00 mg/g at 303 K. On the other hand, that the Pseudo-Second Order model provided the best description of the kinetic uptake properties. The results indicate that FAC is an effective adsorbent for MB adsorption for uptake in dye effluent wastewater streams.

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TABLE 4. Pseudo-First Order (PFO) and Pseudo-Second Order (PSO) adsorption parameter applied to experimental data for the adsorption of MB by FAC at ±303 K

Concentration (mg/L)	Pseudo-First Order			
	qe, cal	*qe, exp*	k_1	R^2
30	9.437	21.292	0.049	0.999
50	1.675	33.046	0.051	0.983
100	36.811	42.922	0.125	0.901
150	21.409	48.858	0.032	0.965
200	23.280	52.055	0.032	0.904
250	25.526	54.795	0.032	0.890
300	30.235	60.731	0.019	0.952
350	28.222	64.384	0.016	0.944
Concentration (mg/L)	Pseudo-Second Order			
	qe, cal	*qe, exp*	k_2	R^2
30	21.882	21.292	0.0063	0.989
50	33.113	33.046	0.0002	0.999
100	51.814	42.922	0.0035	0.955
150	50.000	48.858	0.0017	0.999
200	53.192	52.055	0.0016	0.999
250	56.180	54.795	0.0015	0.999
300	62.112	60.731	0.0019	0.999
350	65.360	64.384	0.0016	0.999

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