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CADMIUM ADSORPTION ONTO NAOH ACTIVATED PALM KERNEL SHELL CHARCOAL

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ABSTRACT: This research aimed to convert the charcoal produced from palm kernel shell pyrolysis into activated carbon nanoparticles for use in the adsorption of cadmium, which is one of the most toxic (e.g., carcinogenic) heavy metals. In particular, the adsorption capacity, adsorption isotherm, and adsorption kinetics of cadmium onto the produced charcoal were determined. Pyrolysis of palm kernel shells at 380°C resulted in liquid smoke, tar, and charcoal. The charcoal was ground with a ball mill until it reached nanoparticle size. Subsequently, it was chemically activated by soaking it in a 0.1 N NaOH solution for 24 h. The pre- and post-activation charcoal was characterized using a scanning electron microscope (SEM). The carbon activated nanoparticles were then used in cadmium adsorption experiments at various contact times (30, 60, 90, 120, 150, and 180 min), several initial cadmium concentrations (30, 60, 90, 120, and 150 ppm), and two stirring speeds (100 and 150 rpm). The results showed that maximum adsorption capacity was reached at 22 mg/g with more than 97% adsorption efficiency. Two adsorption models, i.e., Langmuir and Freundlich, were used to interpret the equilibrium data. At the 100-rpm stirring speed, the Langmuir isotherm indicated a maximum adsorption capacity (qm) for cadmium of 22.37 g/mg (Kl = 0.131 mg/g, with a constant correlation of R^2 = 0.915). The Freundlich isotherm gave values of n = 1.503, Kf = 2.877 mg/g, and R^2 = 0.969. Therefore, these two models are considered suitable for describing the equilibrium cadmium adsorption isotherm in the presence of NaOH-activated palm kernel shell charcoal.

Keywords: Cadmium, Adsorption Capacity, Isotherm, Palm Kernel Shell, Activated Carbon

1. INTRODUCTION

Aquatic and terrestrial heavy metal contamination can lead to serious human and environmental health issues [1,2]. Even at very low concentrations, heavy metals cause concern owing to their mobility and toxicity [3]. One of the most dangerous heavy metals is cadmium. Wastewaters from the electroplating, paint and pigment, battery, metal processing, distillation, pesticides, and plastics industries and household waste are some of the main sources of cadmium contamination. The World Health Organization has set the maximum permissible limit for cadmium in drinking water at 0.003 mg/L [4]. Cadmium is nondegradable and can accumulate within living organisms. Cadmium toxicity causes chronic diseases such as erythrocyte destruction, nausea, salivasi, diarrhea, muscle cramps, kidney degradation, chronic lung problems, and bone deformities [5,6], together with its carcinogenic traits [7]. Therefore, the treatment of cadmium before the wastewater is released into the environment is crucial. Methods for treating cadmium in wastewater include sedimentation, ion exchange, electrochemical technologies, membrane filtration, reverse osmosis, and adsorption [8]. However, these methods have several drawbacks such as their inability to completely separate out cadmium, the length of time they require, and the possibility of forming toxic sediment during the process [9,10]. Previous researchers have stated that cadmium can be treated via the adsorption method using biomass such as pen shells [8], Barbula lambarenensis [11], sunflower seeds [12], compost [13], sesame seeds [14], Chlorella vulgaris [15], grape seeds [16], macrofungus [17], sorghum [18], iron oxide nanoparticles [19], aluminum nanoparticles [20], biochar from Ipomoea fistulosa [21], soil composite [22], and alkali biochar [23]. Among the previously mentioned techniques, the adsorption method has garnered much research attention in terms of addressing the drawbacks of the other methods as it has proven effective and economical in eliminating heavy metals from waste [24,25]. However, even though (i) the process is highly efficient and flexible and (ii) the adsorbent is reusable, the high price of the adsorbent and activator is an issue. Therefore, chemically-activated charcoal produced from palm shell pyrolysis may be an inexpensive candidate adsorbent for treating cadmium contamination of waste waters. These shells have been utilized for many purposes such as boiler fuel, sources of cellulose, and road filler. Recently, palm shells have been used in high-temperature pyrolysis to produce liquid smoke, which can be widely used.

Nonetheless, the sizeable quantities of residual charcoal produced remain largely unused [26,27]. Some of this residual charcoal can be used to make briquettes [28] but its use as an adsorbent is limited to treating copper contamination [29]. This research aimed to determine the effectiveness of NaOH-activated palm shell charcoal by assessing its adsorption capacity, adsorption isotherm equilibria, (Langmuir and Freundlich), and adsorption kinetics toward cadmium.

2. METHODOLOGY

2.1 Adsorbent Preparation

The raw material (charcoal) is a byproduct of the liquid smoke generated from palm shell pyrolysis at 380°C. The detailed pyrolysis procedure has been reported previously [26–28]. The charcoal was ground using a ball mill for 24 h until a nanoparticle size was achieved. It was then chemically activated by soaking it in a 0.1 N NaOH solution for 1 day. The resulting activated carbon was filtered and washed with distilled water to achieve neutral pH before being dried in an oven at 105°C. The aim of the drying process was to remove any residual water and achieve a constant weight. The activated carbon structure was subsequently characterized using SEM.

2.2 Adsorption Test

The activated carbon nanoparticles were used to adsorb cadmium from a $Cd(NO_3)_2$ solution diluted to several concentrations, i.e., 30, 60, 90, 120, and 150 mg/L. For each solution, 100 mL was placed in a 250 mL Erlenmeyer flask and 1 g of adsorbent was added before the mixture was stirred at a speed of either 100 or 150 rpm for contact times of 30, 60, 90, 120, 150, or 180 min. The filtrate was then separated from the adsorbent using a centrifuge and the cadmium concentration determined using an atomic absorption spectrophotometer (AA-7000, Shimadzu, Japan).

3. RESULTS AND DISCUSSION

3.1 Adsorbent Characteristics

SEM was used to observe particle growth and determine particle size. The results for pre- and post- chemical activation using NaOH can be seen in Fig. 1. Fig. 1(a) shows that the physical morphology of the carbon surface prior to activation had a size of 300 nm to 1 µm. Fig. 1(b) shows that after activation, the particle size was below 500 nm, on average. The micrographs also show that the surface structures before and after activation were different, with activation resulting in a better and more even structure.

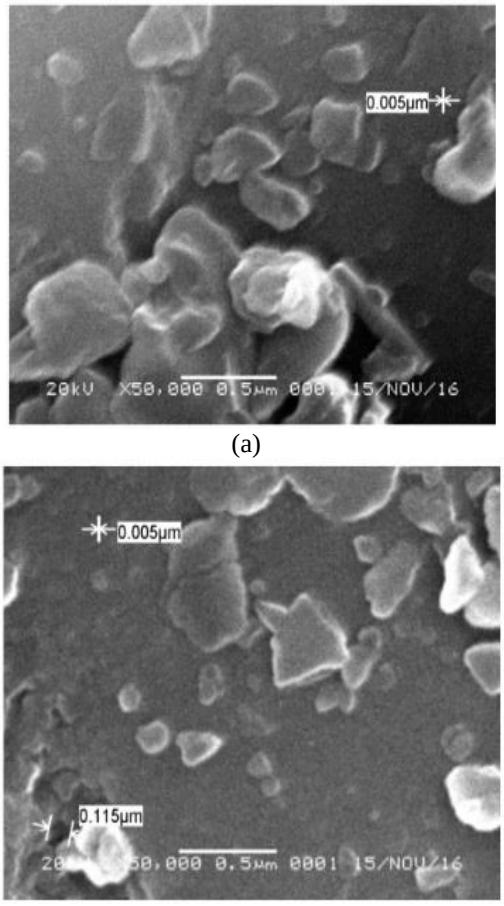


Fig. 1 Physical morphology of the adsorbent surface observed using SEM: (a) before activation, (b) after activation with NaOH

3.2 Study of Adsorption Process

3.2.1 Effect of contact time on cadmium removal

The length of contact time between the adsorbent and the cadmium ions was an important parameter in the adsorption process. Fig. 2 shows that cadmium removal generally increased with increasing contact time, a tendency that was found at all initial concentrations. The highest efficiency was found in the first 30 min, increasing gradually until 150 min. At initial concentrations of 30 and 60 ppm with 30 min contact time, the efficiency rates were 94% and 93%, respectively, increasing to 97% and 96%, respectively. The high efficiencies are likely a result of the contact surface of the adsorbent being relatively large, increasing exposure [30] and adsorption capacity. Longer contact times of up to 150 min resulted in only a slight further increase. In fact, the efficiency started to decrease at 180 min because by then the adsorbent surface had been covered by cadmium molecules and all active sites were saturated. Fig. 3

shows the effect of contact time on the percentage of Cd adsorption at the 150-rpm stirring speed, for which a similar tendency to that observed at 100 rpm was seen. For the initial concentrations of 30 and 60 ppm, cadmium removal was constant up to 60-min contact time, with saturation occurring at 90 min. For the initial concentrations of 120 and 150 ppm, the efficiency continued to increase gradually up to 150 min and then started to decrease at 180 min. Fig.s 2 and 3 show that the highest efficiency was achieved at the 150-min contact time, indicating that 150 min is required to reach equilibrium in terms of cadmium adsorption onto active carbon. Compared with previous research that used an adsorbent made of *Benjamina* leaves extract nano-silver particles, this equilibrium time is relatively high [24].

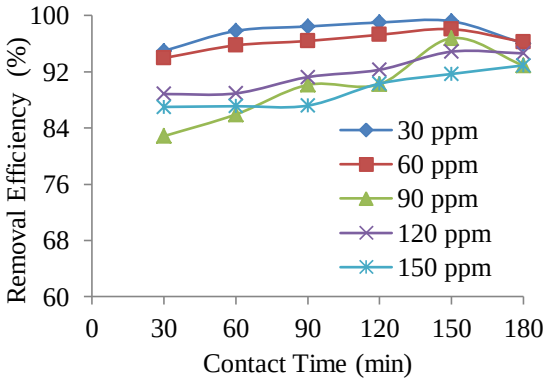


Fig. 2 The effect of contact time on cadmium removal efficiency at 100-rpm stirring speed

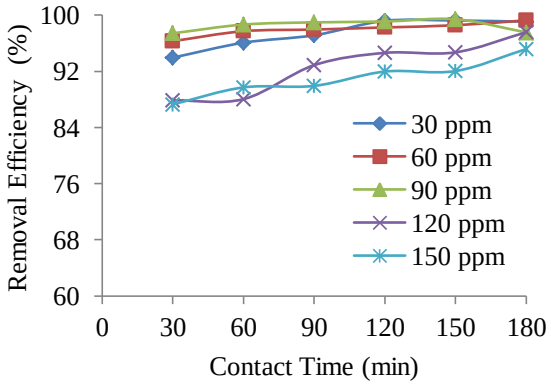


Fig. 3 The effect of contact time on cadmium removal effeciency at 150-rpm stirring speed

3.2.2 Effect of initial concentration on cadmium removal

The effect initial concentration on percentage cadmium removal can be seen in Figs. 4 and 5. Fig. 4 shows that cadmium adsorption efficiency was also influenced by initial concentration.

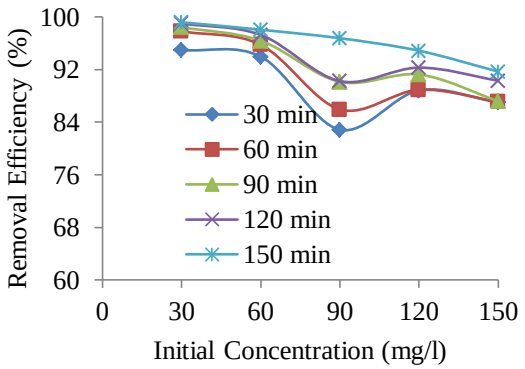


Fig. 4 Effect of initial concentration on cadmium removal at 100-rpm stirring speed

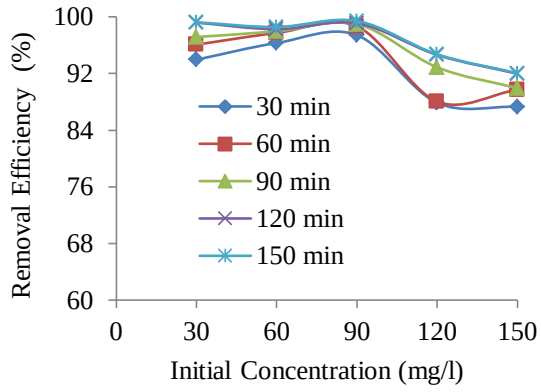


Fig. 5 Effect of initial concentration on cadmium removal at 150-rpm stirring speed

In general, it can be seen that the higher the initial concentration, the lower the adsorption efficiency. For 30-min contact time, the efficiency rates were 93% and 96% for the 30- and 60-ppm initial concentrations, respectively. Further increase in the initial concentration was shown to reduce efficiency, i.e., at 150 ppm, the efficiency decreased to 87%. This is understandable because it essentially means a similar mass of adsorbent had to adsorb more cadmium. The same tendency can be seen in Fig. 5, with optimum adsorption achieved at initial concentrations of 30–90 ppm. This result is similar to that obtained in previous research on dry *Spirulina Platensis* biomass [31]. Stirring speed also affected removal percentage. Slow stirring led to slow adsorption, whereas increased speed likely damaged the adsorbent's structure, preventing it from forming a strong bond with the cadmium ions. Stirring facilitates the transfer of cadmium ions into the active parts of carbon, thereby increasing the mass transfer diffusion between cadmium and an adsorbent's surface [30].

3.2.3 Adsorption isotherm

Analysis of the adsorption equilibrium is essential in determining which model can suitably

describe the adsorption process to be used in the design process. Isotherm equilibrium describes the adsorption and concentration prevalent when equilibrium occurs in a solution. Herein, we used the Langmuir and Freundlich isotherm models to describe this equilibrium, as seen in Figs. 6–9. The adsorption intensity and maximum capacity were calculated from the intercept and slope data. For the Langmuir isotherm, at a stirring speed of 100 rpm, qm = 22.37 and Kl = 0.131 mg/g, which were slightly different from the values obtained at a stirring speed 150 rpm. The Freundlich isotherm constants, Kf and n, were 2.877 and 1.503, respectively. The Langmuir and Freundlich isotherm constants for cadmium ion adsorption can be seen in Tables 1 and 2. Table 1 shows that at a stirring speed of 100 rpm, the correlation coefficients (R^2) for the Langmuir and Freundlich isotherms were 0.915 and 0.969, respectively, whereas at a stirring speed of 150 rpm, the R^2 values were 0.988 and 0.723, respectively. These results show that both isotherm models are suitable for describing the equilibrium of cadmium on NaOH-activated carbon from palm kernel shells. We, therefore, conclude that cadmium ions can be effectively removed from solution owing to a high adsorption capacity [32].

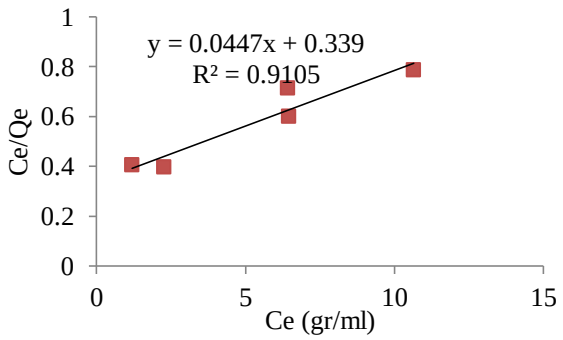


Fig. 6 Analysis of Langmuir isotherm model for cadmium adsorption at 100 rpm stirring speed

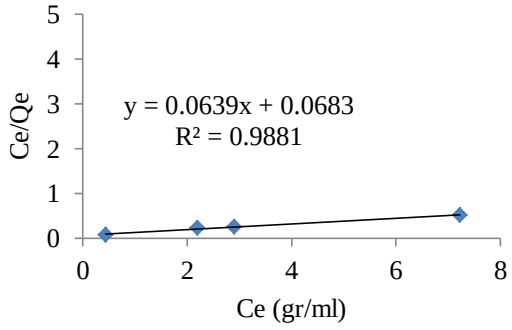


Fig. 7 Analysis of Langmuir isotherm model for cadmium adsorption at 150 rpm stirring speed

Other researchers have found that the Langmuir and Freundlich models are suitable for cadmium adsorption onto chitosan [33], natural phosphate [34], dithiocarbamate-functionalized pyrrole based terpolymers [35], and hydrogel nanocomposites [36]. In addition, the Langmuir isotherm has been reported suitable for describing cadmium adsorption onto maize straw [37] and electronic waste [38]. Moreover, Alimohammadi et al [39] found that the Freundlich isotherm was suitable for describing the use of a nano-adsorbent and magnetic separation to remove cadmium from drilling fluid. The present work shows that the 1/n value for the Freundlich isotherm equilibrium was between zero and one, indicating that cadmium adsorption was preferable at room temperature.

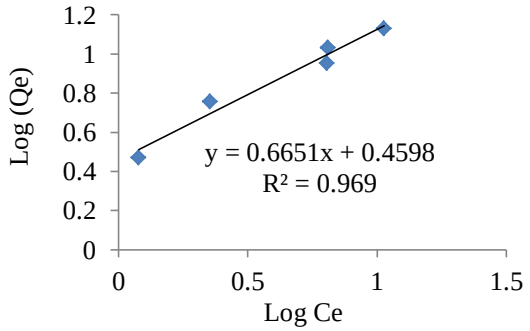


Fig. 8 Analysis of Freundlich isotherm model for cadmium adsorption at 100 rpm stirring speed

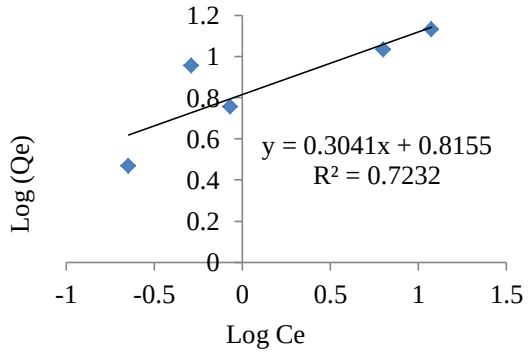


Fig. 9 Analysis of Freundlich isotherm model for cadmium adsorption at 150 rpm stirring speed

In the Langmuir isotherm, the relation between the adsorption process, initial concentration, and adsorption type (whether chemisorption or physisorption) can be determined using the dimensionless separation factor (R):

$$R = \frac{1}{1 + K_L C_i}$$

K_L = Langmuir constant, Co=initial concentration

A value of R between 0 and 1 shows that the adsorption process is favorable; adsorption is linear if R = 1, irreversible if R = 0, and

unfavorable if R > 1 [40,41]. The calculation results are plotted in Fig. 10, showing R values between 0 and 1 for all initial concentrations, indicating that adsorption occurred adequately. However, the value of R declined from 0.202 to 0.0484 at a variety of initial cadmium concentrations from 30 to 150 mg/L. This indicates that adsorption is preferable at a high initial level of cadmium, as reported by Tan et al for phosphoric acid-impregnated carbon [41].

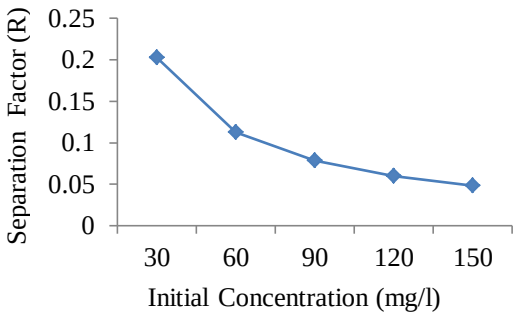


Fig. 10 Relation between R value and initial concentration

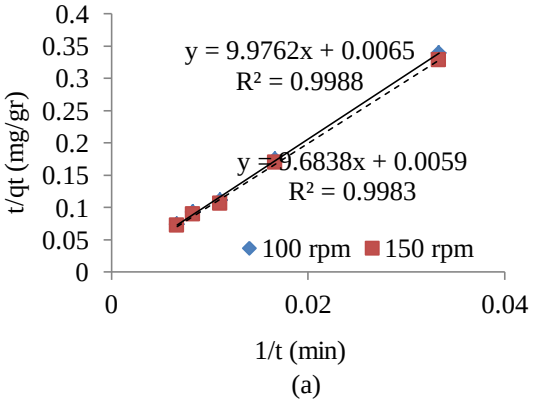
The Langmuir model can be used to calculate the maximum cadmium adsorption capacity, for which we obtained a value of 22.37 mg/g. This value is higher than that found in previous research using sugarcane bagasse, modified carbon nanotube structures, carbon nanofibers, fly ash from heavy fuel, cocoa pod husk biomass, and NaOH-modified sudangrass. Table 3 shows a comparison of the present study with several others that used a variety of adsorbent materials.

Table 3. Comparison of maximum adsorption capacity of various adsorbents for cadmium ions

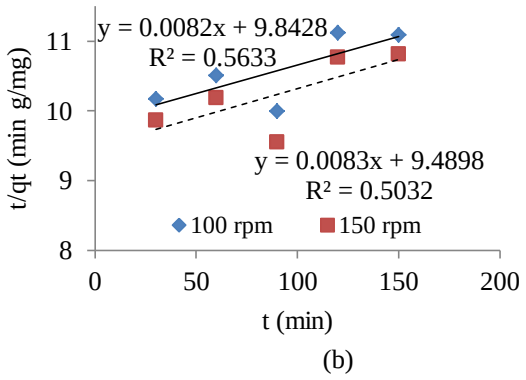
Adsorbent	Maximum adsorption capacity (mg/g)	Reference
Palm kernel shells (NaOH- activated)	22.37	Present study
Pen shells	37.63	Ref.8
Sugarcane bagasse	14.80	Ref.42
Caulerpa fastigiata (Macro algae)	16.13	Ref.32
Banana peel	35.52	Ref.43
Modified carbon nanotube structures (HNO3-activated)	4.35	Ref.30
Carbon nanofibers (HNO_3-activated)	0.935	Ref.30
Activated carbon (HNO_3-activated)	15.9	Ref.30
Fly ash from heavy fuel (HNO_3-activated)	1.55	Ref.30
Cocoa pod husk biomass	13.43	Ref.44
Jatropha seed coat	22.83	Ref.45
NaOH-modified sudangrass	7.76	Ref.18

3.2.4 Adsorption kinetics model

Adsorption kinetics data are important in designing and controlling the adsorption process. This data can be used to optimize cadmium removal performance from industrial waste. The experimental data were regressed to identify the adsorption mechanism using pseudo-first- and second-order reactions. Fig. 11 shows the cadmium ion adsorption kinetics plots for both pseudo-first- and second-order reactions, and Table 4 shows the kinetics parameters for these reactions. For the pseudo-first-order reaction, the values of Qe at stirring speeds of 100 and 150 rpm were 153.84 and 144.92 mg/g, respectively (R^2=0.998 in both cases). For the pseudo-second-order reaction, the Qe values for stirring speeds of 100 and 150 rpm were 21.27 and 40 mg/g, respectively (R^2 = 0.563 and 0.502, respectively). The values of the speed constant for the first-order (K_1) reaction for stirring speeds of 100 and 150 rpm were 1534.76 and 1396.81 g/mg min, respectively. These values indicate that cadmium adsorption onto the active carbon occurred rapidly.



(a)



(b)

Fig. 11 Adsorption kinetics: (a) pseudo-first-order and (b) pseudo-second-order

Table 3 Parameters of pseudo-first-and second-order kinetics models for cadmium adsorption onto NaOH-activated carbon

	Pseudo-first-order			Pseudo-second-order		
	Qe (mg/g)	K_1 (g/mg min)	R^2	Qe (mg/g)	K_2 (g/mg min)	R^2
100 rpm	153.84	1534.76	0.99	21.27	$2.56x10^{-4}$	0.56
150 rpm	144.92	1396.81	0.99	40	$6.73x10^{-5}$	0.50

The correlation coefficient for the pseudo-first-order kinetics model is higher than that for the pseudo-second-order model, signifying that the kinetics data simulation showed a better match with pseudo-first-order kinetics. This indicates that cadmium adsorption onto the NaOH-activated carbon was physical in nature. Although several studies have shown that many adsorbents can be described by the pseudo-second-order kinetics equation, both carbon nanofibers activated with HNO_3 and fly ash have been shown to match pseudo-first-order kinetics, with R^2 values of 0.908 and 0.907, respectively [30].

4. CONCLUSION

NaOH-activated charcoal residues obtained from the pyrolysis of oil palm kernel shells were successfully used to adsorb cadmium from solution, with a relatively high removal efficiency of above 97%. The cadmium adsorption process was optimum at lower initial concentrations; indeed, an increase in the initial concentration up to 150 ppm decreased the adsorption efficiency to 87%. Isotherm studies showed that the Langmuir and Freundlich models were suitable for equilibrium analysis, with R^2 values of 0.915 and 0.969. The maximum adsorption capacity based on the Langmuir model was 22.37 mg/g. Kinetics studies showed that the pseudo-first-order reaction was more suitable for describing cadmium adsorption onto the NaOH-activated carbon, with the following values obtained: Qe = 153.84 mg/g, K1 = 1534.76 g/mg min, and R^2 = 0.998. Therefore, NaOH-activated carbon produced from palm kernel shell charcoal appears to be an acceptable alternative adsorbent for removing cadmium from industrial waste.

5. ACKNOWLEDGMENTS

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