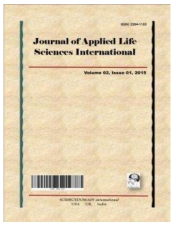

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Activated Carbon Derived from Egyptian Banana Peels for Removal of Cadmium from Water

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Authors' contributions

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

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ABSTRACT

This study was conducted to investigate the removal of cadmium (II) from aqueous solution by using low cost, natural and eco-friendly biosorbent of Banana peels Activated Carbon (BPAC) through batch experiments. The Activated Carbon was characterized by FTIR, SEM, TEM with EDX. Various adsorption parameters such as effect of pH, amount of the biomass, initial concentration of Cadmium ion and contact time on the adsorption capacity of the biosorbent were studied. Biosorption experiments were carried out at the temperature of 25ºC. The percent removal of Cadmium ions was 98.35% from synthetic wastewater. These results were observed at the optimum dose of 0.8 g at pH 4.0. The removal of Cadmium ions from wastewater is mainly influenced by pH of the synthetic wastewater and the amount of biosorbent dose. The equilibrium adsorption data were fitted to Langmuir adsorption isotherm model and the model parameters were evaluated. The kinetic study showed that the pseudo-second order kinetic model better described

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the biosorption process. These results demonstrated that the Banana peels could be used as a natural biosorbent for removal of Cadmium from aqueous solution.

Keywords: Banana peels; activated carbon; cadmium (ii); removal, isotherm; kinetics.

1. INTRODUCTION

The presence of heavy metals in wastewater and surface water is becoming a severe environmental and public health problem [1]. The concentration of these metals in wastewater may therefore rise to a level that can be hazardous to human health, livestock and the aquatic environment [2].

Cd (II) is the one of the toxic metals and has received attention. The removal of cadmium, is gaining wide interest from both environmental and economical viewpoints due to its serious hazardous impacts on humans, animals, and plants. This heavy metal has resulted in serious contamination of both soil and water. Cadmium has been classified as a human carcinogen and teratogen impacting lungs, kidneys, liver and reproductive organs [3]. The World Health Organization (WHO) has set a maximum guideline concentration of 0.003 mg /L for cadmium in drinking water [4].

There are several industries that are responsible for polluting the environment with high level of cadmium ions. The major sources of cadmium are products of industries such as metal plating, cadmium–nickel batteries, phosphate fertilizers, mining, pigments, stabilizers, metallurgy, ceramics, photograph textile printing, lead mining, sewage sludge, alkaline batteries, the combustion of fossil fuels, and electroplating [5].

Therefore, it is important to remove those toxic heavy metals from wastewater. Although heavy metal removal from aqueous solutions can be achieved by conventional methods, including chemical precipitation, oxidation/reduction, electrochemical treatment, filtration, ion exchange and membrane technologies, they may be ineffective or cost-expensive [6]. Recently, adsorption technology has become one of the alternative treatments in either laboratory or industrial scale. There are many adsorbents in use. Activated carbons are known as very effective adsorbents due to their highly developed porosity, large surface area, variable characteristics of surface chemistry, and high degree of surface reactivity [7].

There are several different types of agricultural by-products that have been used for activated carbon (AC) production at laboratory or at an industrial level. Coconut shells [8], walnut shells, olives [9] and apricot stones [10].

Banana (Musa spp.) is considered as one of the most important favorable and popular fruits in Egypt and all over the world. In Egypt, the total cultivated areas reach 5500 feddans which produce about 1.100.000 tons of banana fruits with an average of 28.4 thousand tons/ feddan according to the latest statistic of FAO [11]. Bananas are rich in antioxidant vitamins (vitamins C, A, and E), calcium (Ca), magnesium (Mg), and potassium (K). bananas are considered nutritive with high content of vitamins A and C but poor in vitamins B. Bananas are used fresh or processed into many products such as chips, powder, jams, juice, bar, biscuits, wine etc [12]. At present, these peels are not being used for any other purposes and are mostly dumped as solid waste at large expense. It is thus significant and even essential to find applications for these peels as they can contribute to real environmental problems [13]. Peels are the major by-products obtained during the processing of various fruit and some studies show that these are good sources of polyphenols, carotenoids and other bioactive compounds which possess various beneficial effects on human health [14]. Potential applications for banana peel depend on its chemical composition. Banana peel is rich in dietary fiber, proteins, essential amino acids, polyunsaturated fatty acids and potassium [15].

The utilization of agro-wastes as adsorbents is currently receiving wide attention because of their abundant availability and low cost owing to relatively high fixed carbons and presence of porous structures.

Ball milling is a simple and effective physical method that may increase the total surface area of carbonaceous materials.

The objectives of the present study were to the Preparation of activated carbon from agricultural waste (Banana peels) in nano scale using ball milling (as physical technique to get nano size and pore materials) for the removal of cadmium from aqueous solution. Characterization of the activated carbon was investigated. The equilibrium and kinetic data of the adsorption was studied to describe the adsorption process.

2. MATERIALS AND METHODS

All the chemicals and reagents are analytical grade without further purification. Distilled deionized water was used throughout this work. Materials used in this work were HCl, NaOH and $Cd(NO_3)_2$ purchased from Merck.

2.1 Preparation of the Biosorbent

Banana wastes were collected from local market in Egypt. The waste peels of Banana fruits were first washed with double distilled water and dried in an oven at 80ºC overnight. The dried adsorbent was thermally activated at 500ºC in a muffle furnace for 1h in the presence of air. After activation, the ash contents were removed by washing with distilled water and dried in an oven at 110ºC overnight [16]. The adsorbent Banana peels activated carbon (BPAC) will be ground in nano scale using mechanical ball mill RWTCH Planetary Ball mills type (RM400). Banana Peels is selected for the adsorption experiments are mainly due to its eco-friendly nature and its rich carbon content.

2.2 Preparation of Metal Ion Solutions

A stock solution (1000 mg/L) of Cd(II) ion was prepared by dissolving analytical grade substance of $Cd(NO_3)_2$ in double distilled water. The stock solutions was acidified to desired pH using concentrated HCl in order to prevent the formation of metal hydroxide and to return the metal ion to the dissolve state [17]. Further, the desired pH values of the working solutions were adjusted using 0.1 N NaOH and 0.1 N HCl.

2.3 Batch Biosorption Process

Biosorption studies were conducted in 250 mL Erlenmeyer flask containing a 100 mL of Cd(II) ion. Different amounts of biosorbent; 0.1, 0.3, 0.5, 0.8 and 1.0 g of biosorbent was treated with 20 mg/L of initial metal ion concentration. The flasks were agitated at 200 rpm constantly shaking at a rate of 200 min to establish the equilibrium condition. The samples were taken at definite time intervals at (5, 10, 20 30, 60, 90 and 120 min) and were filtered immediately to remove biomass with filter paper (Whatman 42). The amounts of metal ion were analyzed using Shimadzu AAS 6200 instrument with air–acetylene flame system. The biosorption experiments were carried out at 25ºC. The pH of the test solution was monitored by using a Hanna pH Instruments (Italy).

2.4 Data Analysis

The amount of metal ion adsorbed per unit mass of the biosorbent was evaluated by using the following equation:

$$q_e = \frac{(C_0 - C_e)V}{W} \tag{1}$$

Where C_o is the initial metal ion concentration in mg/L, and C_e is the metal ion concentration at equilibrium in mg/L and V is the volume of metal ion solution in liters, W is the amount of adsorbent used (g). The percent of metal ion removal was evaluated from the equation:

$$\mathrm{Re}\,moval(\%) = \frac{(C_0 - C_e)}{C_0} x100 \tag{2}$$

2.5 Characterization of Activated Carbon

FTIR was used to identify functional groups responsible for metal binding. BPAC and Cd-loaded BPAC (filtered and dried after contact with Cd(II) solution) were mixed with KBr at a ratio of 1:100 and compressed into films for FTIR analysis using a PerkinElmer 1720.

The surface morphology of Banana peels activated carbon (BPAC) was performed by a Scanning Electron Microscopy (SEM) (JEOL 5400), at 30 KV accelerated voltage. Prior to scanning, each adsorbent was coated with a thin layer of gold using a sputter coater to make it conductive. A TEM sample was prepared by dropping diluted solutions of nanoparticles onto 400-mesh carbon – coated copper grids with the excessive solvent immediately evaporated. TEM at an operating voltage of 200 KV determined the morphology and electron diffraction pattern of the nanoparticles. In order to elucidate the biosorption nature of BPAC, the analyses of energy dispersive spectroscopy (EDX) (Jeol, JSM-5800) were used.

3. RESULTS AND DISCUSSION

3.1 Characterization of Adsorbents

3.1.1 FTIR analysis

The FTIR is an important technique to qualitatively determinate characteristic functional groups, which make the adsorption behavior possible.

Fig. 1 shows the FTIR spectra of the prepared nano banana (BPAC). The bands in the region of 3742 and 3642 cm^{-1} are corresponding to phenolic OH stretching bands at 2951 and 2890 cm^{-1} indicate C-H stretching,1697 cm^{-1} indicate C=O and peak around 1570 cm^{-1} can be ascribed to C-C stretching vibration in aromatic rings. The absorption at 1010 cm^{-1} indicates the existence of C–O stretching vibrations in alcohols, phenols, acids, ethers or esters [18]. The weak band in the region of 832 cm^{-1} is attributed to amine groups. Comparing the spectra before (Fig. 1(a)) and after (Fig. 1(b)) sorption of Cd (II), differs in the position of the absorbance peaks appeared. The stretching vibration of hydroxyl group peaks at 3742 cm^{-1} and 3642 cm^{-1} after the biosorption of Cd (II) is significantly disappeared suggesting that chemical interactions occur between the metal ions and the hydroxyl groups on the biosorbent surface. Shifting was observed on the C=O band (1697 cm^{-1} to 1587 cm^{-1}). The prepared nano banana constituted by carbohydrates, proteins, lipids, and fibers were recorded to identify functional groups which responsible for the Cd(II) metal ion coordination.

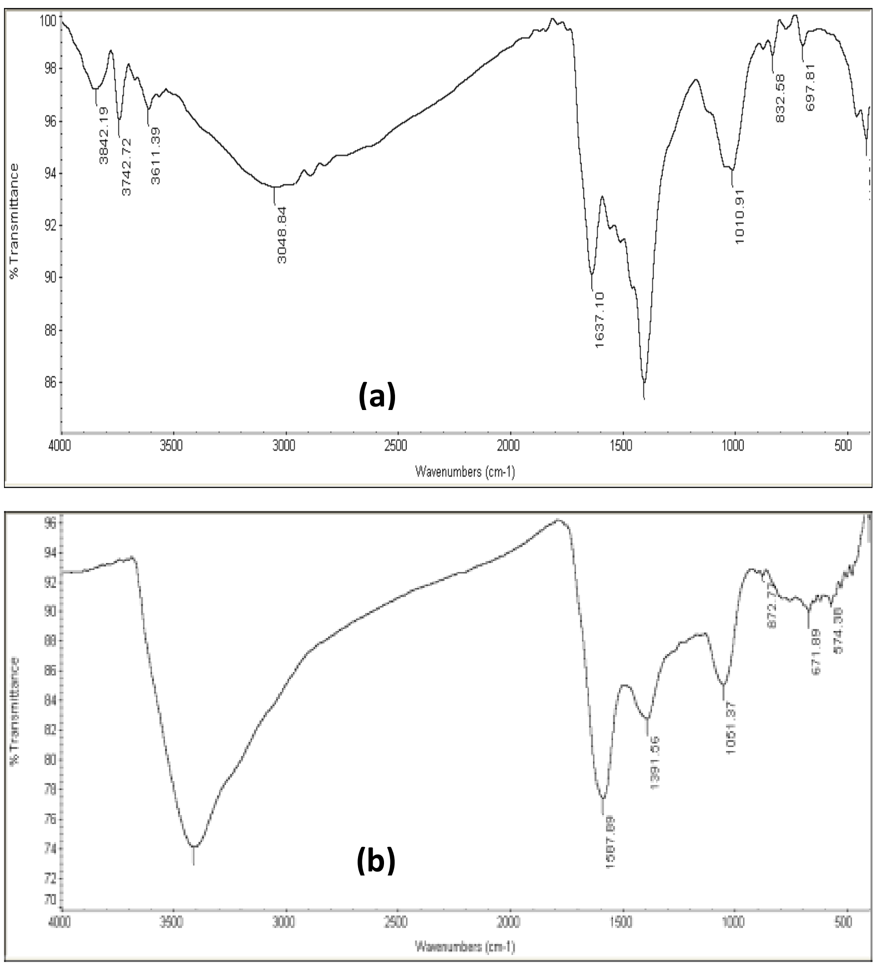

Fig. 1. FTIR spectrum of (a) BPAC before adsorption and (b) after Cd(II) Loaded BPAC

3.1.2 Morphology of the prepared BPAC Nanoparticles

A scanning electron microscope (SEM) was used to examine the surface of the prepared Banana Peels Activated Carbon (BPAC) adsorbent.

Fig. 2 shows the SEM photograph of the prepared activated carbons obtained from BPAC with 500x magnifications

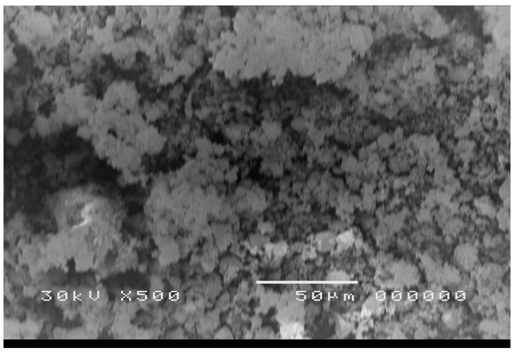

Fig. 2. Scanning electron microscope (SEM) for BPAC

There are many pores available on its surface whereas the external surface of banana has the pores with different sizes and different shapes. The relatively smooth external surface of banana nanoparticles suggests that they are composed of countless tiny primary nanoparticles. The aggregation of these tiny primary nanoparticles can produce large numbers of intra-aggregated pores, resulting in a high microporous volume (as shown in Fig. 2). It is clear that, prepared activated carbons appear to have numbers of pores where, there are good possibilities for Cd(II) ions to be trapped and adsorbed into these pores.

Fig. 3(a) presents the TEM image of the BPAC nanoparticles, BPAC appeared as slightly spherical shape and ranged from 25 to 60 nm in diameter. The selected area of the electron diffraction patterns (SAED) of the prepared sample, show sharp spots that are indicative of single crystalline materials Fig. 3(b).

3.1.3 Energy dispersive spectroscopy analyses (EDX)

In order to know the composition of banana peels, elemental analysis was done with the use of EDX analysis. The EDX analysis is illustrated in Fig. 4, which shows the presence of various elements along with a high amount of potassium, calcium beside carbon and oxygen.

3.2 Effect of pH

The pH is an important parameter influencing the adsorption behavior of adsorbate onto biosorbent surface due to its impact on both the surface binding-sites of the biosorbent and the metal ion solution. In the present study, the effect of pH on biosorption of Cd(II) onto BPAC was studied in a range of 1–6. Results are shown in Fig. 5. It was observed that, the removal of Cd(II) ions in the aqueous solution is low at low pH values and gradually increased with increasing pH of the solution up to pH 4.0. With further increase in pH, there is no significant increase in the amount of metal ion removed. Maximum removal is observed at pH 4.0. Hence, all further experiments were carried out at pH 4.0. Similar findings have been reported by other researchers working on removal of heavy metal ions from aqueous solution by activated carbon prepared from Tunisian date stones [19].

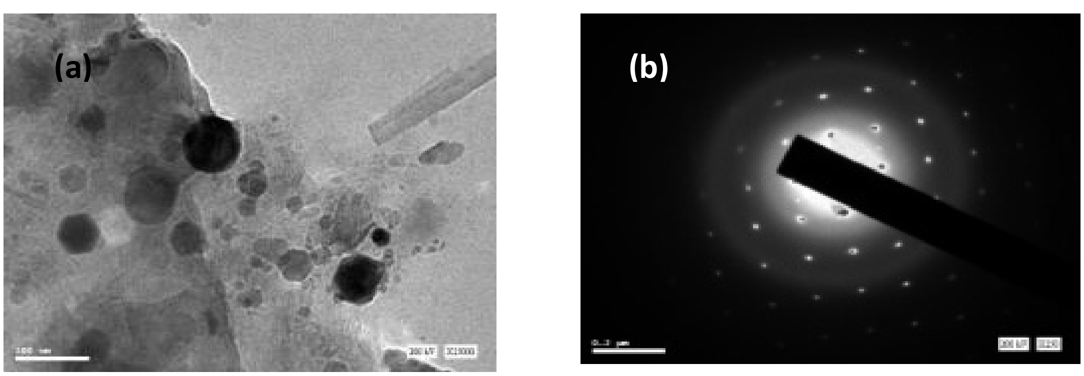

Fig. 3(a). Transmission electron microscopy (TEM) of BPAC, (b) electron diffraction patterns (SAED) of BPAC

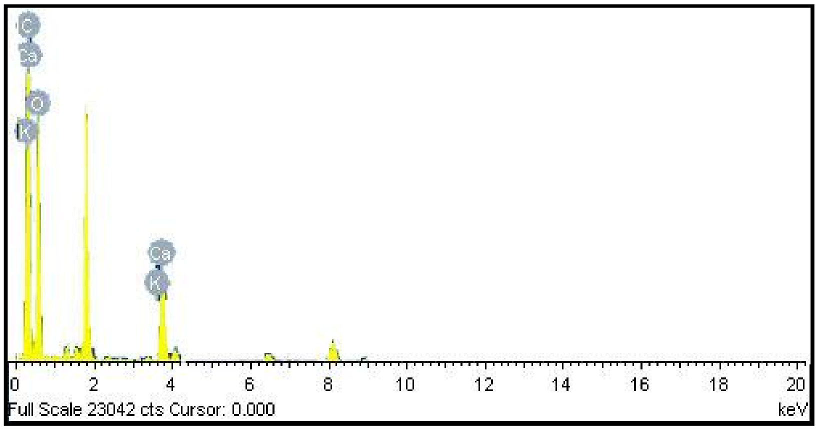

Fig. 4. EDX analysis of BPAC

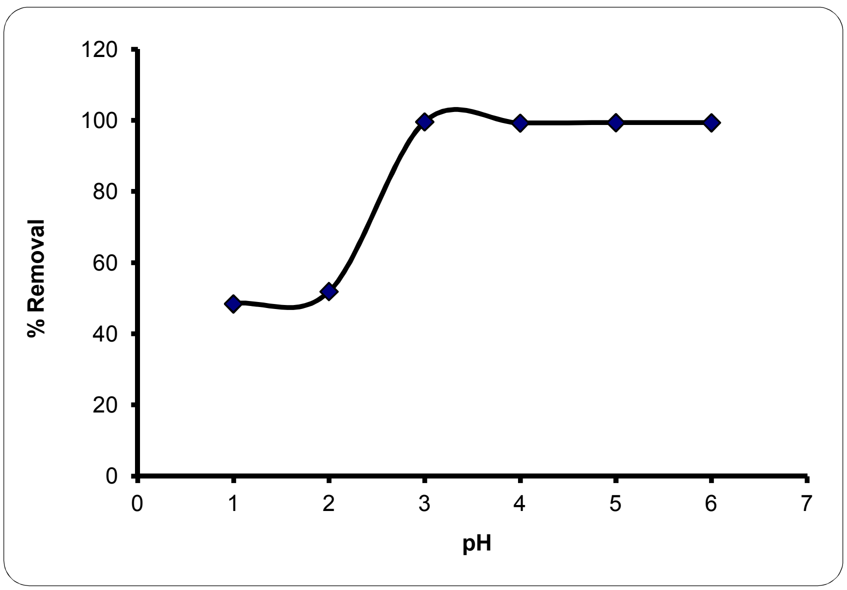

Fig. 5 . Effect of pH on biosorption of Cd(II) by BPAC, adsorbent dose 0.8 g, initial metal ion concentration 20 mg/ L, temperature 25ºC

At low pH values, concentration of H^+ ions far exceeds that of the metal ions and hence H^+ ions compete with Cd(II) ions for the surface of the adsorbent which would hinder the Cd(II) ions from reaching the binding sites of the adsorbent resulting in low adsorption amount of Cd(II). As the pH increases, there are fewer protons in the solution and consequently there is lesser competition with Cd(II) for binding sites [20].

3.3 Effect of Adsorbent Dose

The dependence of metal ions on effect dose was studied by varying the amount of biosorbent from 0.1 to 1.0 g/100 ml, while keeping other parameters (pH, contact time, and initial concentration of metal ion) constant. As it can be seen in Fig. 6, the percentage removal of Cd(II) increases with increase in the dose of adsorbent from 0.1 to 1.0 g. There was a non-significant increase in percentage removal of Cd(II) when adsorbent dose increases beyond 0.8 g. This suggests that after a certain dose of biosorbent, the maximum adsorption is attained and hence the amount of ions remains constant even with further addition of dose of adsorbent. This is due to an increase in the surface area of the biosorbent, which in turn increases the number of binding sites. However, at high sorbent doses the available metal ions are insufficient to cover all the exchangeable sites on the biosorbent, resulting in low metal uptake [21,22]. Similar trends were observed in different adsorbents [23,24]. Similar behavior for the effect of biosorbent dose was observed for biosorption of Manganese ion (Mn^{+2}) on Egyptian Banana Peel [25].

3.4 Effect of Contact Time

The biosorption efficiency of Cd(II) ions was evaluated as a function of contact time. The initial concentration of Cd(II) ions was 20 mg/ L. The relationship of percentage Cd (II) ions removal by BPAC with contact time is shown in Fig. 7, which shows that the biosorption increased until the contact time reached 90 min. After 90 min., the biosorption efficiency was almost constant such that it could be considered the equilibrium time of the Cd(II) biosorption. To ensure that sufficient contact time was obtained, further biosorption experiments were carried out for 90 min.

3.5 Effect of Biosorbent Concentration

The effect of the concentration of BPAC was studied using 100 mL solutions of Cd(II) with different biosorbent concentrations ranging from 10 to 100 mg/L. The other parameters were kept constant. The experimental results are presented in Fig. 8. The biosorption capacity of BPAC increased from 1.241 to 12.487 mg/g with increasing of the initial Cd(II) concentration. It is well know that the removal efficiency of metals depend on the type and quantity of the biosorbent. An increase in biosorption concentration generally increases the biosorbed Cd(II) ion concentration because of large biosorption surface area. The percentage removal of metal ion decreases with increase in initial metal concentration and shows little decrease at higher concentrations. This can be explained by the fact that the adsorbent has a limited number of active sites that become saturated at a certain concentration [26]. Although the percentage removal of metal ion decreases, the equilibrium biosorption capacity of the adsorbent increases with increasing metal ions concentration. The initial metal ion concentration provides an important driving force to overcome all the mass transfer resistance between the solution and solid phases, hence a higher initial concentration of metal ion may increase the biosorption capacity.

3.6 Adsorption Isotherms

An adsorption isotherm represents the equilibrium relationship between the adsorbate concentration in the liquid phase and that on the adsorbents surface at a given concentration. A number of isotherms have been developed to describe equilibrium relationships. The data obtained from biosorption of Cd(II) ions were analyzed using Langmuir and Freundlich isotherms. The Langmuir model suggests that uptake occurs on a homogeneous surface by monolayer sorption without interaction between the adsorbed molecules [27]. The linear form of Langmuir adsorption isotherm is given by the following equation;

$$\frac{C_e}{q_e} = \frac{1}{Q_m b} + \frac{C_e}{Q_m} \qquad (3)$$

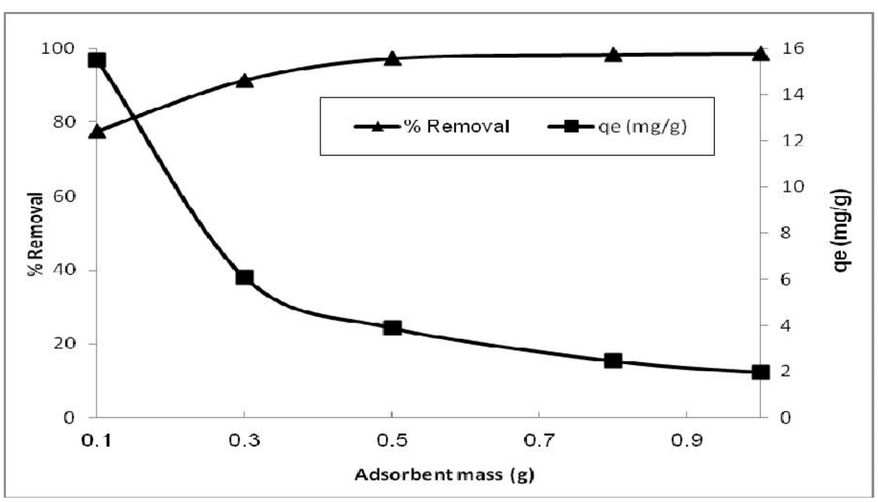

Fig. 6. Effect of biosorbent dose of Cd(II) by BPAC (experimental conditions: initial Cd concentration = 20 mg/ L, pH 4.0, agitation speed = 200 rpm, contact time = 2 h at 25ºC

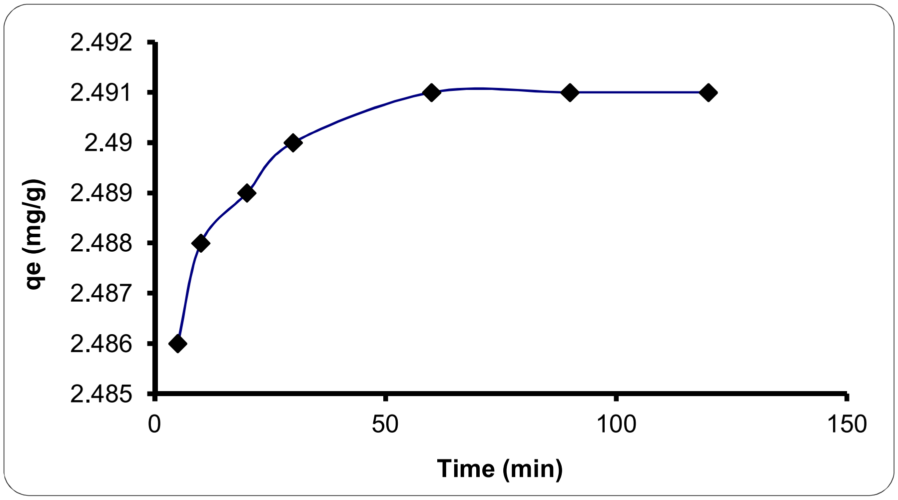

Fig. 7. Effect of contact time on Cd(II) adsorption on BPAC, pH = 4.0, adsorbent dose 0.8 g, agitation speed = 200 rpm, metal ion concentration: 20 mg/L)

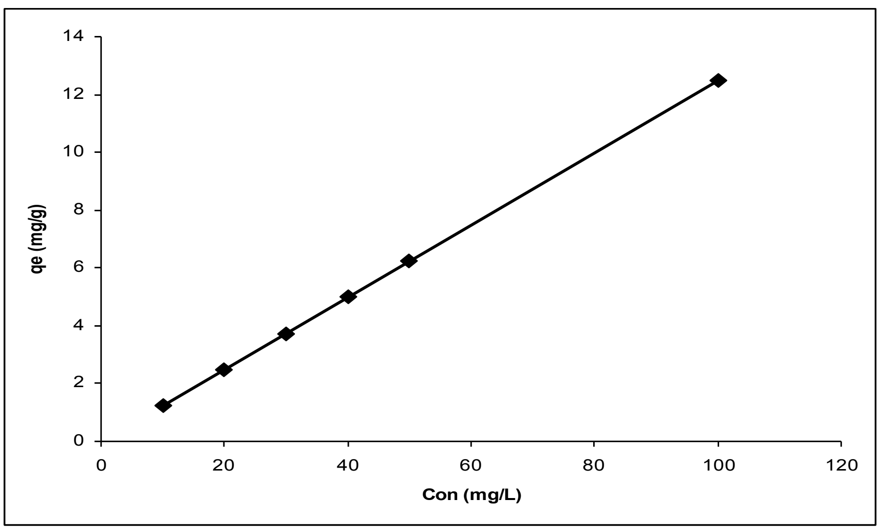

Fig. 8. Effect of initial metal ion concentrations of Cd(II) biosorption on BPAC, adsorbent dose 0.8 g, agitation speed = 200 rpm, temperature 25ºC

Where q_e is the equilibrium metal ion concentration on the biosorbent (mg/g), b is the Langmuir constant and q_m is the monolayer adsorption capacity. The plot of C_e /q_e versus C_e is employed to generate the intercept value of $1/bq_m$ and slope of $1/q_m$ (Fig. 9).

One of the essential characteristics of this model can be expressed in terms of the dimensionless separation factor for equilibrium parameter, R_L, defined as:

$$R_L = \frac{1}{1 + bC_0} \quad (4)$$

The value of R_L indicates the type of isotherm to be irreversible (R_L=0), favourable (0< R_L<1), linear (R_L=1) or unfavourable (R_L>1). The value of R_L in the present study was found study was found to be 0.0008 indicating that the adsorption of Cd(II) on BPAC is favorable.

The adsorption data were analysed using Freundlich adsorption isotherm [28]. The linear form of the Freundlich adsorption isotherm is;

$$\log q_e = \log K_f + 1/n \log C_e \quad (5)$$

Where n is an empirical parameter which

reflects the intensity of adsorption that varies with the degree of heterogeneity and K_f is a constant related to adsorption capacity. The application of Freundlich isotherm to the data of biosorption of Cd(II) onto BPAC (data not shown) indicated the inapplicability of the model judged by low correlation coefficient.

It is found that Langmuir isotherm model fit very well according to the correlation coefficient values given in Table 1. Thus, the results of the present study indicate that biosorption of Cd(II) onto BPAC is homogeneous in nature.

Table 1. Adsorption isotherms for removal of Cd(II) ions by BPAC

Biosorbent	Langmuir isotherm parameters		
(BPAC)	q_m (mg/ g)	*b* (L m/g)	R^2
	0.719	10.526	0.9973

3.7 Kinetic Studies

For analyzing the adsorption kinetics of cadmium ions, the pseudo-first- and pseudo-second-order were applied to the experimental data. The pseudo first order rate equation is one of the most widely used equations for the adsorption of a solute from an aqueous solution and is represented as:

$$\log(q_e - q_t) = \log(q_e) - \frac{K_1}{2.303}(t) \qquad (6)$$

Where q_e and q_t are the amount of metal ion adsorbed (mg/g) at equilibrium and time t, respectively. K_1 is the first-order reaction rate constant (l/min). Examination of the data shows that the pseudo-first order kinetic model is not applicable to Cd(II) adsorption onto BPAC judged by low correlation coefficient.

3.8 The pseudo-second-order

The linear form of the pseudo-second-order kinetics rate is given in the following equation:

$$\frac{t}{qt} = \frac{1}{K2qe2} + \frac{1}{qe}(t) \qquad (7)$$

Where q_e is the equilibrium biosorption capacity and K_2 is the pseudo-second order rate constant (g/mg.min). Pseudo second order model assures that the rate of occupation of adsorption sites is proportional to the square of the number of unoccupied sites [29]. A plot of (t/q_t) versus t gives a linear relationship for the applicability of the second-order kinetic model (Fig. 10).

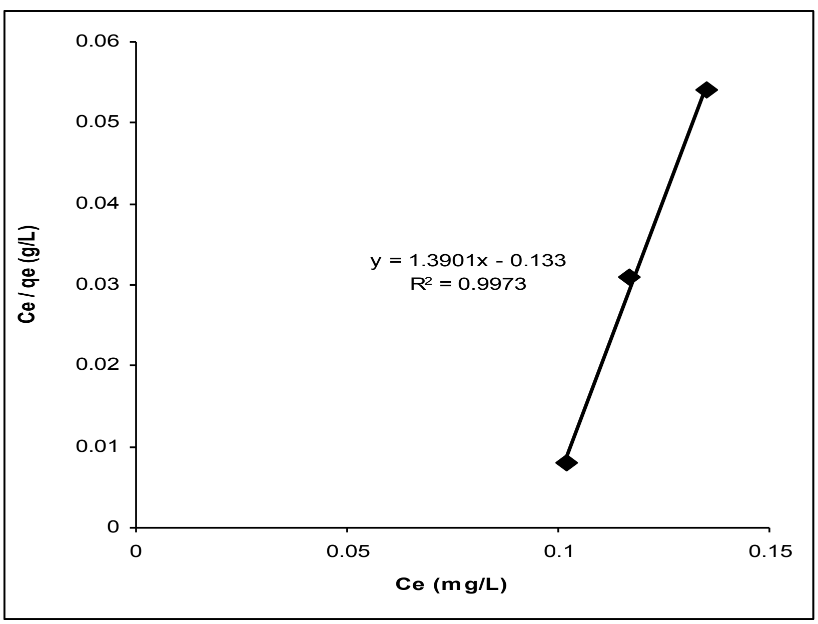

Fig. 9. Langmuir isotherm plot for adsorption of Cd(II) onto BPAC

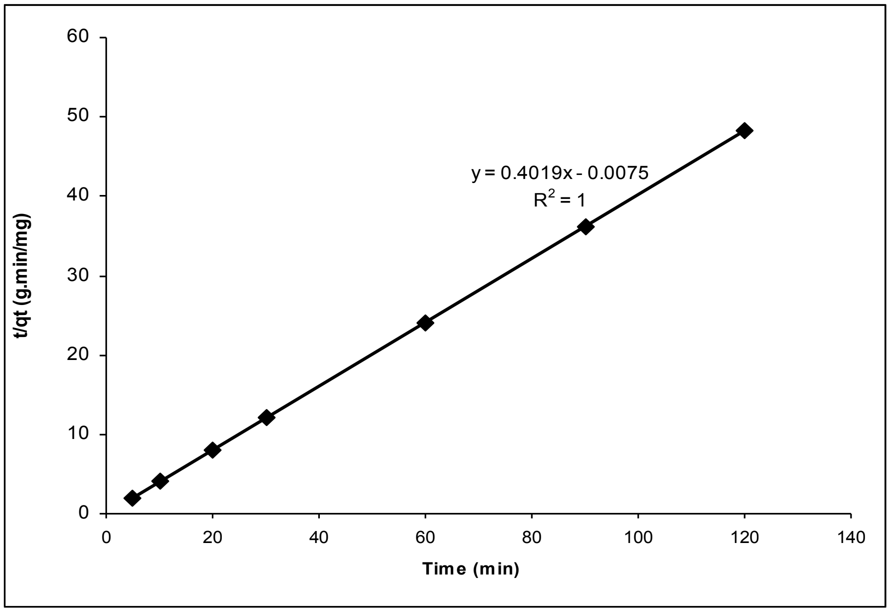

Fig. 10. Pseudo-second order kinetic plot for the biosorption of Cd(II) onto BPAC at 25ºC

As seen from the Table 2, due to high R^2, for the pseudo second- order model better for Cd(II) ion than the first order model. The result indicates that, the removal of cadmium from the synthetic wastewater is well followed the pseudo second order kinetics.

Table 2. Kinetic parameters for the removal of Cd(II) ions by BPAC

Kinetic model	Parameters	
Pseudo-second order	q_e (mg/g)	2.488
	K_2 (g/mg.min)	21.551
	R^2	1.000

4. CONCLUSION

This study investigated the removal of cadmium by Banana Peels Activated Carbon (BPAC) from an aqueous solution. The experimental results showed that maximum pH for efficient Cd(II) ions biosorption was about 4.0. In this batch mode of studies, the adsorption was dependent on pH, contact time, initial metal ion concentration and biosorbent dose. The biosorbent was successful in removing Cd(II) with 98.35% adsorption efficiency from the aqueous solution containing 20 mg/l Cd(II). The biosorption of Cd(II) onto BPAC was well fitted with the Langmuir isotherm model. The pseudo-second order model expressed the biosorption kinetic well. This study revealed that the BPAC can be used as an economical natural based biosorbent to remove Cd(II) from aqueous solution. Because the BPAC can be obtained as cheap and easily available waste material in many countries and used for future commercial application.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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