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Research Article

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Cadmium Removal from Aqueous Solutions Using Dried Banana Peels as An Adsorbent: Kinetics and Equilibrium Modeling

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Abstract

The use of Dried banana peels as an adsorbent for removal of cadmium ions from aqueous solutions has been studied. Batch experiments have been conducted at different concentrations to evaluate the maximum adsorption capacity of Dried banana peels. The influence of pH, contact time, adsorbent dose was investigated at room temperature. Langmuir and Freundlich adsorption isotherm models used to test equilibrium of adsorption. The process of adsorption was found to be fast and equilibrium has been reached in within 2 hours. The maximum adsorption capacity of cadmium on Dried banana peels is 5.91 mg/g, evaluated by Langmuir adsorption isotherm model. Pseudo-first-order and pseudo-second-order kinetic models applied to evaluate rate constants. FTIR spectra of adsorbent showed the presence of hydroxyl, carboxylic and amine groups in dried banana peels. This study shows that banana peels has great potential for removal of cadmium ions and can be used as a good adsorbent for removal of cadmium from water and wastewater at very low concentration.

Keywords: Adsorption; Cadmium ions; Banana peels; Langmuir isotherm; Kinetic study

Introduction

Heavy metals are considered as primary pollutants due to its toxicity and mobility in natural water system. Among the various heavy metals, cadmium is considered as extremely toxic and carcinogenic to human beings. Cadmium is non-biodegradable heavy metal persists in environment for a long time causing harmful effects to aquatic ecosystem even at a very low concentration in water [1,2]. In natural water bodies, the major anthropogenic sources of cadmium contamination that include discharge of wastes and effluent from industries such as metallurgical processes, electroplating, plastics manufacturing, battery manufacturing, pesticide and fertilizer industry and mines [3,4]. Past events like Itai-Itai disease at Toyama Prefecture in Japan showed the extent of health effects due to the contamination of cadmium in water [5]. The cadmium contamination also causes the health effects for instance high blood pressure, bone fraction, destruction of RBCs, reproductive toxicity, hepatic effects and immunological effects [6,7]. The permissible limit of cadmium in drinking water is 0.003 mg/L as guided by World Health Organization (WHO) [8].

Several treatment technologies are available for removal of cadmium from water like chemical precipitation, reverse osmosis, ion exchange, membrane filtration, electrolysis etc. [9] but these treatment technologies have found to be expensive, high chemical demanding and least efficient to remove cadmium at low initial concentration. In recent years, biosorption is found to be a promising method than that of other methods as it is non-expensive, easily operational, regenerable and ecofriendly method [10-13]. Removal of cadmium at low initial concentration has wide applications for treatment of cadmium contaminated water and can effectively acts as a finishing treatment in effluent treatment plants to reduce cadmium level bellow discharge limits of stipulated standards.

Various types of agricultural waste materials like rice husk, sugarcane bagasse, saw dust, brazil nutshell, grape stack, mango peels and coconut copra meal etc. are studied as an adsorbent for removal of cadmium and other heavy metal ions from water and waste water [5,12,14]. In literature, it is reported that functional groups like carboxyl, hydroxyl, phosphate, thio and amino present on the walls of agricultural waste biomass play an important role for binding of heavy metals. These functional groups bind metal through ion exchange by transferring hydrogen ions or through complex formation by sharing electron pair [15]. Most of these agricultural materials showed significant adsorption capacities than Granular Activated Carbon [13]. Cadmium removal by using banana peels has been studied previously in multicomponent system [10] and they achieved adsorption capacity of 5.71 mg/g for cadmium removal onto dried banana peels.

The present investigation is undertaken to study the removal of cadmium as a single metal from aqueous solutions by using Dried banana peel as an adsorbent. Banana is one of the major fruit consumes worldwide in large quantity. Discarded peels of banana fruit from market area and household garbage generates waste in bulk quantity. Banana peels constitutes cellulose, hemicelluloses, lignin and pectin in its biomass containing functional groups like carboxyl, hydroxyl and amine. These functional groups are reported as important for binding of metal ions on biosorbents [14,16,17].

In this study batch experiments have been carried out for optimization of various process parameters like initial pH, contact time, adsorbent dose and initial metal ion concentration. The maximum adsorption capacity of Dried banana peels has been evaluated by using Langmuir adsorption isotherm equilibrium and intensity of adsorption was evaluated by Freundlich adsorption isotherm. Kinetic study conducted by using pseudo-first-order and pseudo-second-order kinetic models. The intraparticle diffusion model is applied to test whether intraparticle diffusion is rate limiting factor or not. The adsorbent material was characterized by FTIR spectroscopy to identify

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affinity of functional groups present in adsorbent material for removal of cadmium ions.

Materials and Methods

Preparation of adsorbent material

Banana peels from Banana fruits was collected from local market of Nagpur, India. Banana peels were dried in sunlight for 3 days and cuts into small pieces then washed with distilled water and dried it in oven at 70°C for 5 hours. Oven dried pieces were grounded and sieved using American standard 60 mesh sieve.

Preparation of the stock solution of cadmium nitrate

Stock cadmium solution (1000 ppm) was prepared by dissolving of dried analytical grade cadmium nitrate Cd $(NO_3)_2.4H_2O$ (Sigma-Aldrich) of ≥ 99% purity into double distilled water. Working solutions of concentrations 1, 5, 10, 15, 20, 25, 30, 40, 50 ppm were prepared by using appropriate amount of stock cadmium solution.

Apparatus

The concentration of cadmium was measured by using Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) (Thermo Scientific iCAP 6300) at a wavelength of 228.8 nm at 0.5 ml/min pump speed. Argon gas was used to generate plasma. pH meter-1500 (Eutech Instrument) was used to adjust the pH of solution. FTIR spectrum of adsorbent material was analyzed by Bruker Vertex 70 FT-IR Spectrometer (2:100 adsorbent: KBr ratio). Remi Orbital Shaker was used for agitation of the solutions.

Batch study for process parameters

Parameters selected for study was contact time, adsorbent dose, pH, initial concentration. Effect of these parameters was studied by keeping one parameter constant and alternatively changing other parameters.

The amount of cadmium adsorbed onto dried banana peels was calculated by using mathematical mass balance equation.

$$q_e = \frac{(c_0 - c_e)\,v}{m} \tag{1}$$

Where, q_e is amount of cadmium adsorbed on adsorbent; c_0 is the initial metal ion concentration in solution; c_e is the metal concentration in solution at equilibrium; v is volume of solution in liter and m is the mass of adsorbent in grams.

Study of adsorption isotherm

To study adsorption isotherm, various concentrations of cadmium 5, 10, 15, 20, 25, 30, 40 and 50 mg/L were prepared by using cadmium stock solution (1000 mg/L). Initial pH of solution was in the range of 2-3 and adjusted to 6-7 using 0.1 N HCl or NaOH solution. 0.5 gm of Dried banana peels added to each flask containing 100 ml cadmium solution. Whole content was agitated on orbital shaker for the time of 120 min at room temperature, 25°C. During agitation, flasks were covered with aluminum foil to avoid evaporation. After completion of contact time, contents of flask were filtered through whatman filter paper No. 41 and filtrate was analyzed for the residual cadmium concentration in solution by using ICP-OES instrument. Recovered adsorbent was kept for drying for further material characterization.

The Langmuir and Freundlich adsorption isotherm models describes well the equilibrium between cadmium ions adsorbed onto the surface of adsorbent and residual metal ion concentration in solution at constant temperature [10,18].

Langmuir adsorption isotherm: Experiments carried out at various initial concentration ranging from 5 ppm to 50 ppm at constant temperature of 25°C. Langmuir adsorption isotherm plotted by using standard straight line equation which considers monolayer coverage of adsorbent. Monolayer adsorption capacity was evaluated using equation [19];

$$\frac{1}{q_e} = \frac{1}{b.q_m.c_e} + \frac{1}{q_m} \tag{2}$$

Where q_e (mg g^{-1}) is the amount of metal adsorbed, and c_e (mg L^{-1}) is concentration at equilibrium, q_m (mg g^{-1}) is the maximum adsorption capacity of adsorbent and b (Lg^{-1}) is the Langmuirs constant. The graph of $1/c_e$ *vs.* $1/q_e$ was plotted to evaluate the values of q_m and b from the slope and intercept of graph.

Freundlich adsorption isotherm: Freundlich adsorption isotherm equations were used to model adsorption behavior of cadmium by Dried banana peels. Freundlich (Eq. 3) adsorption isotherms was plotted by using standard straight-line equation and corresponding two parameters K_F and n was calculated from graph of log c_e verses log q_e [20].

$$\log qe = \log K_F + \frac{1}{n}\log c_e \tag{3}$$

qe (mg g^{-1}) is the amount of metal adsorbed, and c_e (mg L^{-1}) is concentration at equilibrium K_F and n are Freundlich isotherm parameters which indicates adsorption capacity and adsorption intensity, respectively.

Study of adsorption kinetics

It is important to know the rate of batch sorption kinetics for the optimization of contact time and for design of adsorption unit in water treatment system. Kinetics experiments were carried out using 250 ml conical flasks. Into each flask 100 ml of solution was taken which spiked with cadmium (II) concentration to 1 ppm and 10 ppm. Then added 0.5 g Dried banana peels to each flask and kept for shaking on mechanical orbital shaker at shaking speed of 85 rpm. Contact time of different time interval (5 to 200 min) were given upto reach equilibrium. After specific contact time, the whole content was filtered through whatman filter paper No. 41 and separated adsorbent from solutions. Filtrate was analyzed for residual cadmium concentration.

All the experiments were carried out at 25°C. A blank solution without adding adsorbent was processed for same process in all the experiments to correct experimental errors. All the experiments were carried out for three times, out of which two identical readings were taken into consideration.

Results and Discussion

Fourier transform infrared (FTIR) spectroscopy of adsorbent

The FTIR spectra of biosorbent revealed that Dried banana peel contains number of functional groups in its biomass indicating complex nature of biosorbent. Two separate samples of adsorbent i.e., raw adsorbent and cadmium loaded adsorbent were analyzed for FTIR. Spectra of adsorbent (Figures 1 and 2) shows transmittance peaks at various frequencies indicated presence of different functional groups. In the raw adsorbent, strong peak due to O-H stretching at the frequency 3485, and 3365 cm^{-1} indicated that the existence of free hydroxyl group of polymeric compounds such as lignin or pectin that containing the functional groups of alcohols, phenols and carboxylic acids. A broad range of frequency (3600 to 2800 cm^{-1}) is assigned to free hydroxyl group indicating the presence of polymeric compounds [21,22]. The

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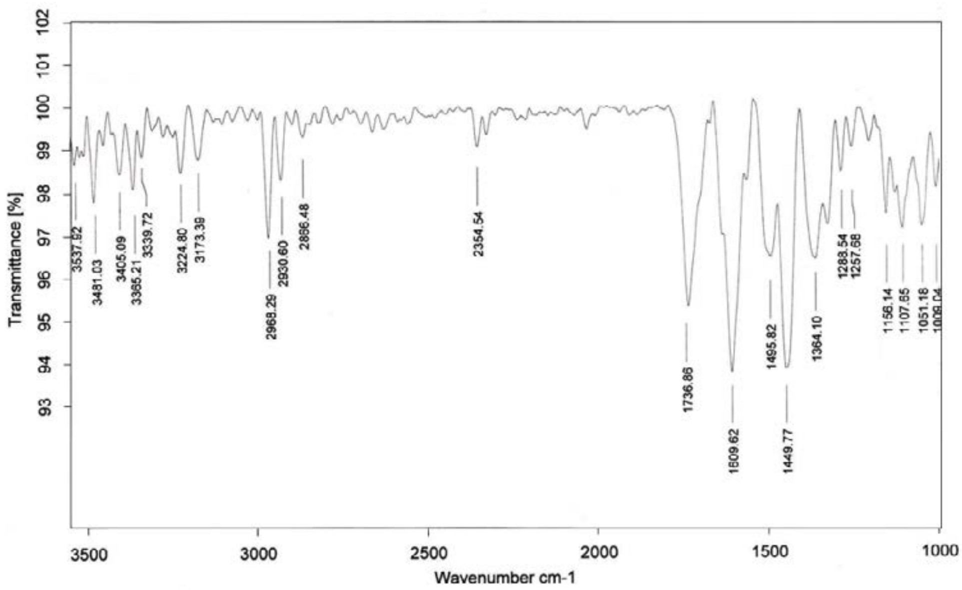

Figure 1: FTIR spectra of Dried banana peel.

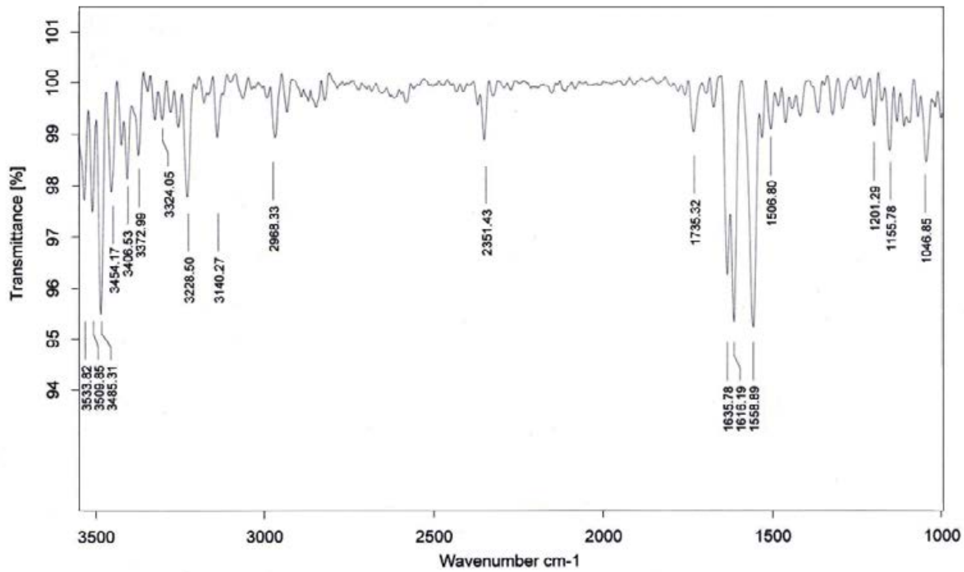

Figure 2: FTIR Spectra of Cadmium loaded Dried banana peel.

peak due to N-H bending vibrations of primary amines was observed at 1609 cm^{-1}. A peak at 1736 cm^{-1} is due to C=O stretching vibrations of carboxylic groups (-COOH, -$COOCH_3$) which can be attributed to carboxylic acids or their esters [23]. Difference in transmittance at these peaks of cadmium loaded adsorbent after adsorption indicated that carboxylic group is involved in binding mechanism. The peak at 1465cm^{-1} may be due to aromatic ring vibration of lignin. Peaks at the wave number 1288 and 1364 cm^{-1} can be attributed to C-H bending of crystalline cellulose and C-H bending of cellulose, hemicelluloses or lignin polymer and the peak at 1107 cm^{-1} assigned to stretching vibrations of C-N bond of aliphatic amines. FTIR spectra of raw and cadmium loaded Banana peel indicate that Banana Peels composed functional groups like hydroxyl, carboxyl and amine groups. Lowering in transmittance and small deflection in band frequencies after adsorption at these peaks shows the participation of these functional groups in adsorption [14,23,24].

Effect of adsorbent dose

The adsorption of cadmium was studied with increasing of adsorbent quantity of adsorbent in the range of 0.1 to 3 g/100 ml (Figure 3). Result

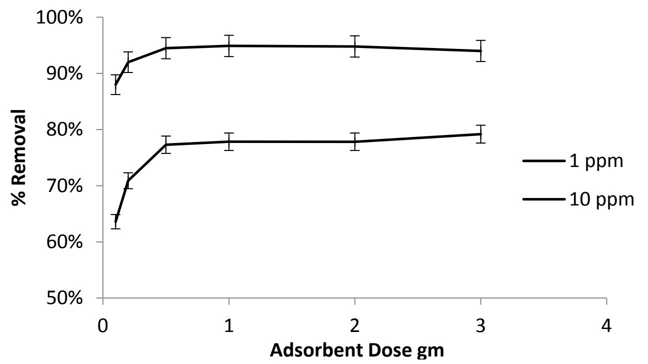

Figure 3: Effect of adsorbent dose on cadmium uptake (contact time, 120 min; agitation speed, 85 rpm; temperature, 25°C).

showed that adsorption efficiency of banana peels increased with increase of adsorbent dose. Increasing adsorbent dose makes available more active surface sites for binding of cadmium. Maximum removal was observed at adsorbent dose of 0.5 g/100ml. Increase in adsorbent dose beyond optimum quantity of 0.5 g/100 ml, the removal percent of cadmium not increased considerably because of reduction in available effective surface area [25].

Effect of pH

pH is one of the most important factors affecting adsorption process for binding of cadmium. pH of solution studied in the range of 1-10 (Figure 4). In acidic pH range cadmium ions compete with H^+ ions for binding on available surface sites therefore lower adsorption was observed in the pH range of 1 to 4. Highly increase in rate of metal removal was observed at pH range of 4-7. At pH greater than 4 deprotonation of functional groups like carboxylic group (-COOH) may occur [17]. It makes available more negatively charged active sites on adsorbent. It results into strong attraction of metal ion on surface of adsorbent. In alkaline pH range above 8, the adsorption seems to be better and greater removal was observed. In alkaline pH range precipitation of metal hydroxides may occur. Metal removal may not involve complete adsorption in this pH range therefore pH range 7-8 was selected for further adsorption study [17,26].

Effect of contact time and shaking speed

Contact time is also one of the most important factors affecting the adsorption of cadmium. The effect of contact time was evaluated for 1 ppm and 10 ppm initial cadmium concentration at different time interval (5-200 min) by keeping pH, adsorbent dose and shaking speed constant. The increased adsorption of cadmium was observed with increasing of contact time. The result showed that maximum adsorption (85% removal) of cadmium was occurred at first 10 min of contact time. Adsorption rate gradually increased up to of 50 min of contact time. After 50 min less increase in adsorption rate was observed and remained almost constant after 120 min (Figure 5). Less increase in adsorption rate after 50 min may be due to lowering of driving force for adsorption as less difference between initial concentration and solid liquid interface occur after 50 min [27]. Optimum contact time for cadmium adsorption was selected as 120 min for further experiments. To prevent settling of adsorbent material at bottom of flask and to give more contact with maximum active adsorption surface, optimum shaking speed of 85 rpm was observed.

Adsorption Isotherms

Langmuir adsorption isotherm: The sorption data of different initial concentration was well fitted into Langmuir adsorption isotherm with R^2 value 0.99 (Figure 6). The maximum adsorption capacity q_m (mg/g) for cadmium was found to be 5.91 mg/g. The Langmuir's parameter 'b' indicated bond energy for binding of metal with adsorbent material by complexation reaction. The essential characteristics of Langmuir adsorption isotherm can be explained by dimensionless separation factor 'r'. The value of 'r' describes the type of adsorption isotherm as follows;

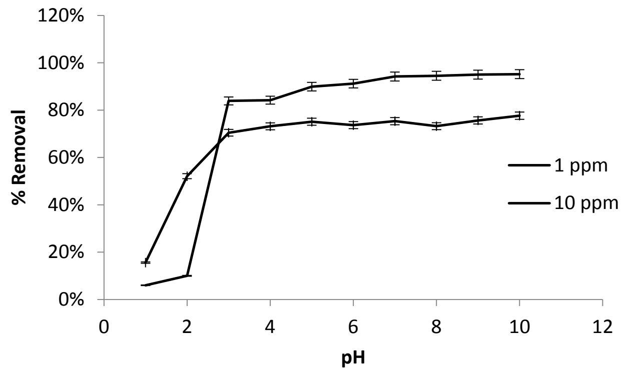

Figure 4: Effect of pH on cadmium removal (adsorbent dose 0.5 g/100 ml; contact time, 120 min; agitation speed, 85 rpm; temperature, 25°C).

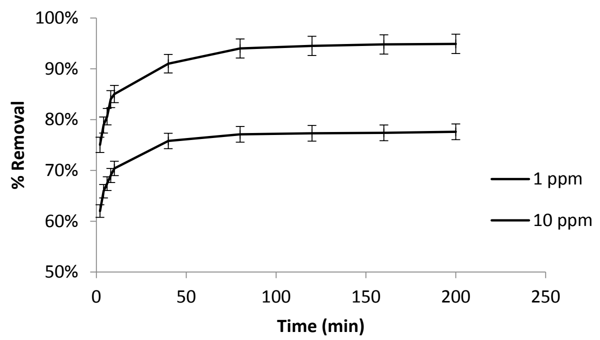

Figure 5: Effect of time on cadmium removal (adsorbent dose 0.5 g/100 ml; agitation speed, 85 rpm; pH, 8; temperature, 25°C).

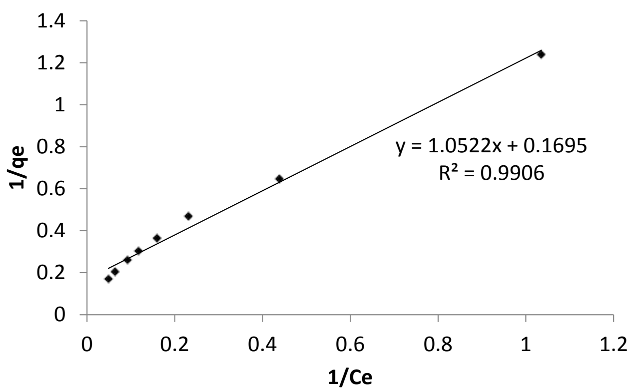

Figure 6: Langmuir isotherm for cadmium (metal concentration, 5-50 mg/L; contact time, 120 min; agitation speed, 85 rpm; temperature, 25°C; pH, 8.).

$$r = \frac{1}{(1+b.c_0)} \tag{4}$$

Where b is the Langmuir's constant and c_o is the initial concentration of cadmium.

For each initial concentration, the calculated value of separation factor 'r' found in the range of 0.573 to 0.118 which lies in between 0 and 1, it indicates that the adsorption is favorable [18,27]. The maximum adsorption capacity of banana peels to adsorb cadmium evaluated from Langmuir adsorption isotherm reported in literature is presented in Table 1. It is observed that banana peels have significant adsorption capacity. The maximum adsorption capacity evaluated by this study is matches the results obtained previously in literature [10].

Freundlich adsorption isotherm: Freundlich adsorption isotherm for cadmium is presented in Figure 7 and the corresponding parameters K_F (the ultimate adsorption capacity) and n are calculated from the graph of log c_e *vs* log q_e. The value of n indicates the measure of adsorption intensity of cadmium on banana peels (Table 2). If the value of n=1, the adsorption is linear; if n<1, the adsorption is a chemical process; if n>1, adsorption is a physical process [12]. The value of n was observed to be 1.57 which indicates the cadmium adsorption onto banana peels involves physical process. The value of R^2 0.99, indicated that isotherm holds good for cadmium adsorption.

Kinetics study

Kinetic study was carried out to examine adsorption capacity of adsorbent and to evaluate the rate constant. Adsorption kinetics describes the rate of solute uptake by adsorbent with increasing contact time of 2, 4, 6, 8, 10, 40, 80, 120, 160 and 200 min. Various types of kinetics models were used in different studies of heavy metal removal using adsorbent but out of that pseudo first order, second order and intraparticle diffusion kinetics models are widely used for the adsorption of metal ions (Figures 8-10). Applicability of models expressed by correlation coefficient (R^2) [18,28,29].

Pseudo first-order-kinetic model: Lagergrens pseudo first-order-kinetic model is used to describe adsorption rate of cadmium based on

Sl No	Material used	q_m mg/g	Reference
1	*P. ruscipolia* wood	7.40	[3]
2	*A. donax* L.	5.70	[3]
3	Brazil nuts shell	19.4	[3]
4	Sugarcane bagasse	10.7	[3]
5	Peels of banana	5.71	[10]
6	Fungal *P.tenuiculus*	11.4	[33]
7	Mango peel waste	68.92	[12]
8	*Raphanus sativus* peels	19.82	[28]
9	*Moringa olifera* bark	39.41	[27]
10	*Nauclea diderichii* seed biomass	6.30	[32]
11	Castor seed hull	6.98	[18]
12	Coconut Copra meal	2.59	[34]
12	Grannular activated carbon	3.70	[18]
13	Dried banana peels	5.92	**Present study**

Table 1: Adsorption capacity of cadmium onto banana peel and other agro based biosorbents.

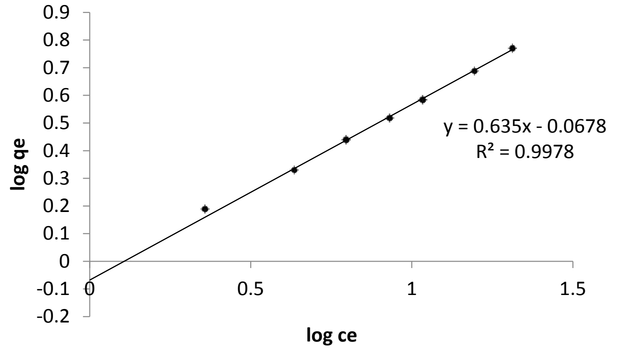

Figure 7: Frieundlich isotherm for cadmium (metal concentration, 5-50 mg/L; contact time, 120 min; agitation speed, 85 rpm; temperature, 25°C; pH, 8.).

Langmuir Isotherm Parameters			
Metal	R^2	q_m (mg g^{-1})	b (L g^{-1})
Cd	0.99	5.92	0.149
Freundlich Isotherm Parameters			
Metal	R^2	n	K_F
Cd	0.99	1.57	0.857

Table 2: Langmuir and Freundlich isotherm parameters.

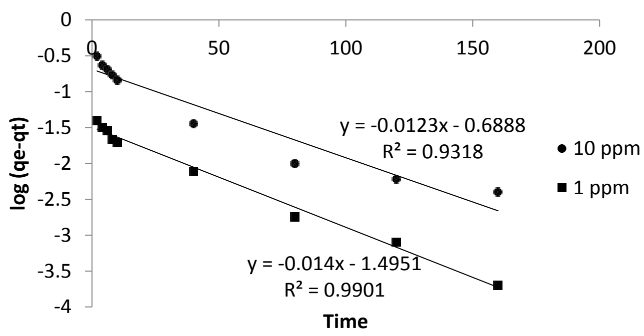

Figure 8: Pseudo first order plot of t *vs.* log (qe-qt).

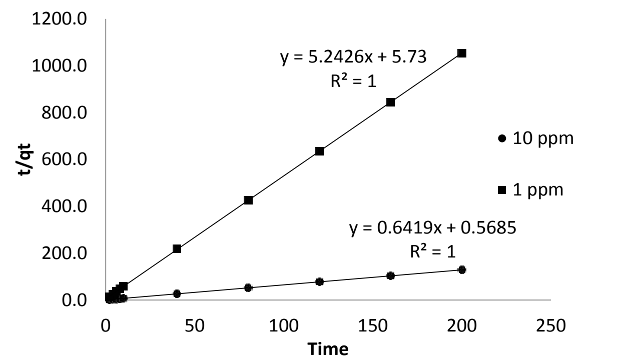

Figure 9: Pseudo second order kinetics plot of t *vs.* t/qt.

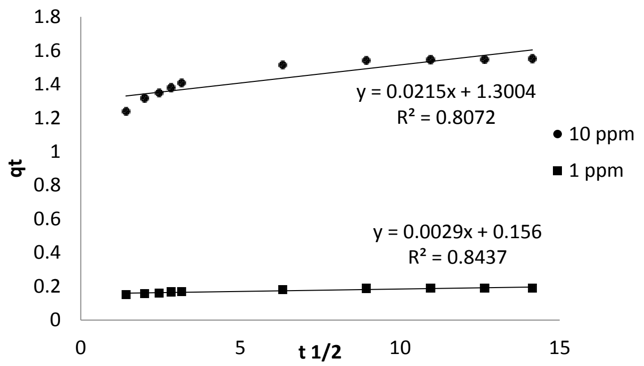

Figure 10: Intraparticle diffusion model: Graph of t1/2 *vs.* qt.

the adsorption capacity of adsorbent. Lagergrens equation is expressed as follows:

$$\frac{dqt}{dt} = k_1(q_e - q_t) \quad (5a)$$

Where, q_e is the amount of cadmium adsorbed (mg g^{-1}) at equilibrium, q_t is the amount of cadmium adsorbed (mg g^{-1}) at any time t (min), k_1 is equilibrium rate constant of pseudo first order adsorption (l min^{-1}). Integrating the equation (5a) for boundary layer conditions t=0 to t=t and q=0 to q=t gives,

$$\log\left(\frac{q_e}{q_e - q_t}\right) = \frac{k_1}{2.303}t \quad (5b)$$

This equation (5b) represents integrated rate law for pseudo first order reaction. The linear form of equation is expressed as follows,

$$\log(q_e - q_t) = \log qe - \frac{k_1}{2.303}t \quad (5c)$$

Where, k_1 is the rate constant. The correlation coefficient (R^2) value calculated from the plot of log (q_e-q_t) *vs.* t is found to be high (Figure 4). But the calculated values of qe from plot not matches with experimental values for both the 10 ppm and 1 ppm concentrations (Table 3) It indicates that adsorption of cadmium onto dried banana peels does not follow first order kinetics.

Pseudo second-order-kinetics model: The linear form of pseudo second order kinetic model is given by equation [30],

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (6a)$$

Where, k_2 (g mg^{-1} min^{-1}) is the rate constant of pseudo second order equation. q_e is the adsorption capacity of adsorbent at equilibrium, q_t is the amount of cadmium adsorbed by adsorbent at time t. The value of q_e and k_2 can be calculated from the slope and intercept of the linear plot of t *vs.* t/q_t which is presented in Table 3. The experimental value of q_e matches to predicted values of q_e from the plot with correlation coefficient value (R^2=1) which suggested that, second order kinetic model fits better to describes the kinetics of adsorption [28].

As experimental data fits, well in second order kinetics (Figure 4), the values of initial sorption rate 'h' and half adsorption time '$t_{1/2}$' can be calculated from the following equations (9) and (10), respectively which are based on equation of second order kinetics.

$$h = k_2 q_e^2 \quad (6b)$$

$$t_{1/2} = \frac{1}{k_2 q_e} \quad (6c)$$

Intraparticle diffusion model

Intraparticle diffusion model proposed by Weber and Morris [31] has been widely used by researchers for the analysis of adsorption kinetics. The process of metal adsorption onto adsorbent mainly involves three steps including bulk diffusion, film diffusion and pore diffusion or intraparticle diffusion. Out of these, intraparticle diffusion process is slow process and it may be a rate limiting process. Adsorption of metals generally controlled by intraparticle diffusion or liquid phase mass transport rates. In this study, experiments were carried out by batch mode with rapid stirring of the contents therefore intraparticle diffusion may be the rate limiting step [29]. Intraparticle diffusion model is applied to test whether it is a rate limiting step or not. The intraparticle diffusion rate constant (k_d) is calculated from the relationship between q_t and the square root of time $t^{1/2}$ is expressed in following equation (7) provided by Weber and Morris [31].

$$q_t = k_d t^{1/2} + B_L \quad (7)$$

Where, K_d (mg g^{-1} $min^{-1/2}$) is intraparticle diffusion rate constant, B_L (mg g^{-1}) is proportional to thickness of boundary layer. The values of K_d and B_L are calculated from the slope and intercept of plot of q_t *vs.* $t^{1/2}$ (Table 3). The plot of adsorption capacity of adsorbent (q_t) versus square root of time ($t_{1/2}$) is found to be linear with R^2 value of 0.80 and 0.84 for 10 ppm and 1 ppm initial concentrations, indicated considerable linearity representing that intraparticle diffusion is involved in adsorption process (Figure 5). The slope of graph (k_d) is 0.021 and 0.002 mg g^{-1} min^{-1} for 10 ppm and 1 ppm initial concentration respectively, indicating that some of boundary layer may be involved in adsorption. If the line of linear graph passing through origin, it indicates that intraparticle diffusion may rate limiting factor but in the graph line is not passing through origin, having intercept value greater than zero [32-34]. It suggested that intraparticle diffusion may not rate limiting factor and surface adsorption along with intraparticle diffusion may play important role in adsorption of cadmium onto dried banana peel [18,28].

Conclusion

Dried banana peels show a good efficiency for the removal of

C_0 mg/L	1ppm	10ppm
Pseudo first-order		
q_e (exp)/mg g^{-1}	0.189	1.552
q_e (cal.)/mg g^{-1}	0.031	0.205
k_1 (min^{-1})	0.032	0.0276
R^2	0.99	0.93
Pseudo second-order		
q_e (cal)/mg g^{-1}	0.190	1.557
K_2 (min^{-1})	4.83	0.641
h (mg g^{-1} min^{-1})	0.174	1.56
$t^{1/2}$	1.08	1.0
R^2	1.0	1.0
Intraparticle diffusion		
B_L (cal)/ mg g^{-1}	0.15	1.30
k_d (g mg^{-1} min^{-1})	0.002	0.021
R^2	0.84	0.80

Table 3: Comparison of various factors at different initial concentration.

cadmium ions from aqueous solution. FTIR spectra of banana peel indicates that the presence of functional groups like hydroxyl and carboxyl. Adsorption capacity evaluated by Langmuir adsorption isotherm is found to be 5.91 mg/g which is slightly greater than previous studies. Experimental data has well fitted into second-order kinetic model suggesting second-order nature of the process. Intraparticle diffusion suggested that intraparticle diffusion is not the rate limiting factor for adsorption. The present study concludes that easily available, nonhazardous agricultural waste material like banana peels can be successfully used as an adsorbent material for removal of cadmium from aqueous solutions even at low concentration.

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