

Adsorptive Capacity, Adsorption Isotherm and Adsorption Kinetics of Activated Carbon for the Removal of Toxic Cadmium from Polluted Water

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Abstract: *Environmental pollution with metals is a serious issue worldwide posing threats to humans, animals & plant. Cadmium is a heavy metal that occurs as a natural constituent in earth crust. Cadmium is toxic to living systems and therefore, it is essential to remove from wastewater. Activated carbon has been used for the adsorption of Cd (II) from solution. Adsorption capacity of activated carbon for Cd (II) abatement was investigated employing batch equilibration method. The effects of various parameters like contact time, pH, particle size and Adsorbent dose have also been studied. The adsorption data were found to fit well with the Freundlich and Langmuir isotherm models. Activated carbon under investigation has been used to be an excellent economical adsorbent material for Cd (II) removal from contaminated water.*

Keywords: Activated carbon, Adsorption, Cadmium, kinetic, isotherm

1. Introduction

Recently, heavy metals in water have become a global problem because most of them are toxic to organisms [1], non-degradable and accumulate in biota. Among these metals, cadmium is one of the most toxic and poses a significant risk to human health because, in excess, it causes serious diseases and many disorders [2-3]. The main sources of cadmium entering the environment come from wastewater discharged from many industrial sectors such as metal plating, Cd-Ni batteries, stabilizers, pesticides, paints, pigments, plastics, metallurgy, textile printing industries and alloys [4-5]. Different methods have been used for cadmium removal, such as adsorption, ultrafiltration, coagulation, reverse osmosis, solvent extraction, electro-dialysis, flocculation and membrane separation [6-9]. These methods are very expensive and are only effective at high levels of metals present in aqueous solution [10]. Among them, the adsorption technique is an inexpensive conventional treatment technology, commonly used in the treatment of wastewater and the removal of heavy metals from the environment, due to its simplicity, short treatment time, ease of use, efficiency, and its ability to recycle the adsorbent and heavy metals [11-12]. The key factors determining the choice of a suitable adsorbent are adsorption capacity, abundant local supply, low price and eco-friendly nature. Due to their metal sequestration properties, natural materials are ecologically relevant for reducing the concentration of heavy metals or dyes present in aqueous media, as demonstrated by the adsorption capacity of biosorbents materials, such as chicken eggshells [13-15], peanut shells [16], tea leaves [17], coffee husks [18], corncob [19] olive pomace [20] and olive seeds [21]. The US EPA has established a maximum contaminant level of 0.005mg/L for Cadmium in drinking water (0.005 ppm). In present study, activated carbon is used for the removal of Cadmium from water and wastewater by batch adsorption technique. Adsorption is considered one of the most effective and low-cost

methods to eradicate heavy metals from water [22]. It is an environmentally friendly technique with flexibility in design and operation. The process of adsorption is reversible and the adsorbents can be regenerated through desorption of the adsorbed species. Meanwhile, different types of biological, agricultural and industrial materials and mineral oxides have been applied with high efficiency for the removal of heavy metals [23- 24].

Over the last few decades, activated carbon has been extensively applied as a potent adsorbent for the expulsion of poisonous metals from wastewater due to the tunable surface chemistry and well-developed permeable structure with a high specific surface area. The selection of activated carbon as an adsorbent is, therefore, extremely significant as heavy metals are removed through complexation or by electrostatic attraction between the metal ions and different oxygen carrying functional groups present on the surface of the activated carbon adsorbents [25-28].

2. Materials and Methods

2.1 Preparation of adsorbent (activated carbon):

Activated carbon was modified by heat treatment [29] where it was washed with deionized water and then heated to 450°C for 4 hrs. The samples were then dried at 100°C and ground to a particle size of a 53 µm to obtain a fine powder of the activated carbon.

2.2 Preparation of adsorbate:

A stock solution (1000 mg/L) of Cd (II) was prepared by dissolving an accurately weighed analytical grade Cadmium sulphate ($3\text{CdSO}_4 \cdot 8\text{H}_2\text{O}$) in de-ionized water. Working solutions of different concentrations, as required, were prepared from the stock by dilution process.

2.3 Batch adsorption experiments:

Batch adsorption experiment were carried out in which 20 cm³ of each metal ion of different concentrations was mixed with a definite mass (0.05 - 0.30 gm) of the adsorbent in 150 cm³ plastics bottles and were agitated in a horizontal mechanical shaker at 140 rpm for a pre-determined period of time and temperature at natural pH. The mixture was filtered through whatmann filter paper, and filtrate analyzed for its residual metal content using atomic absorption spectrophotometer (S- series by Thermo Electron Corporation). All experiments were carried out in duplicate and mean values determine were presented.

The amount of Cd (II) adsorbed q (mg/g) was evaluated thus:

$$q_t = \left[\frac{C_0 - C_t}{M} \right] V \quad (1)$$

$$q_e = \left[\frac{C_0 - C_e}{M} \right] V \quad (2)$$

Percentage removal of adsorbate =

$$\left[\frac{C_0 - C_t}{C_0} \right] 100 \quad (3)$$

Where, q_t and q_e are the amount of Cd (II) adsorbed (mg/g) at time t and at equilibrium respectively; C_0 and C_t are the initial concentration at $t = 0$ and its concentration at time $t = t$ (mg/L); M = the mass of Adsorbent (gm), V = volume of Cd (II) solution (L).

3. Results and Discussion

3.1 Effects of the initial concentration of cadmium (II) ions and contact time:

The effect of contact time on the amount of Cd (II) adsorbed by adsorbent was studied using three different initial concentrations (2, 5 and 10 mg/L) so as to optimize the adsorption time and to study the kinetics of the adsorption process. As contact time increases, the concentration of Cd (II) in the solution decreased rapidly at the beginning and later slow down until it remained constant at about 90 minutes, which was then taken as the equilibrium time (Figure.1). This indicates that the removal of Cd (II) by adsorbent was very rapid at the beginning.

In fact, in all the initial concentrations used, over 70% removal was achieved within the first 10 min. Figure.2 indicates the time course for the percentage removal for the adsorption of Cd (II) by adsorbent. The initial rapid phase may be due to the large number of vacant sites available at the initial period of the adsorption. The amount of Cd (II) adsorbed per unit mass of the adsorbent also increases with increasing initial concentration of the

metal ions. For example, the amount of Cd (II) removed at 40 minutes contact jumped from 0.77 mg/g at the initial concentration of 2 mg/L to 1.78 mg/g and 3.06 mg/g, at the initial concentrations of 5 and 10 mg/L respectively (Figure.3). This may be because an increase in initial concentration enhances the interaction between the ions and the surface of the adsorbent. This, however, leads to a corresponding reduction in the percentage removal of the metal ion (97, 89 and 77% respectively) (Figure.3). Removal efficiency was higher for low concentration because of availability of unoccupied binding sites on the adsorbent. Percentage removal decreased with increasing concentration because at high metal concentrations the binding sites were almost completely covered.

3.2. Effects of adsorbent dose:

The dependence of adsorption of Cd (II) ions on adsorbent dose was investigated by varying the quantity of the adsorbent from 0.02 gm to 0.30 gm in 20 mL of 20 mg/L solution of the ion while keeping other parameters (contact time, agitation speed, particle size, temperature) constant. The results show that the adsorption capacity of adsorbent decreases with increasing adsorbate concentration. However, the removal efficiency of the adsorbent generally improved with increasing dose increased from 93% to 96 % as the adsorbent dosage was increased from 20 mg to 300 mg (Figure.4). This can be attributed to the increase in the adsorbent pore surface areas and availability of more adsorption sites with increasing mass of adsorbent. The increase in removal efficiency with increasing adsorbent concentration was rapid at low dosage but gradually falls and describes a typical saturation curve.

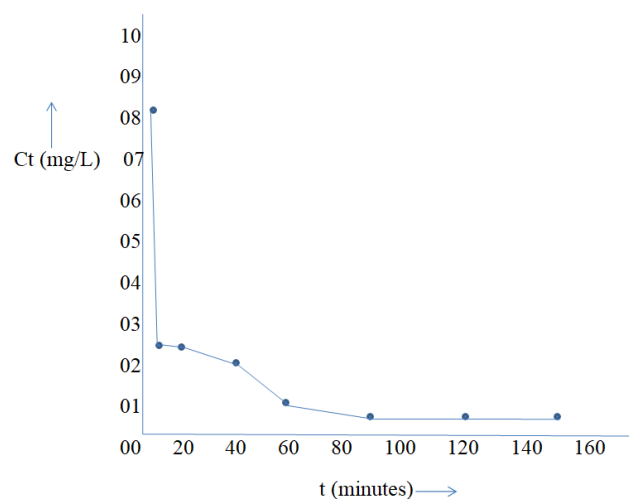


Figure 1: Effect of contact time on the residual concentration of Cd (II)

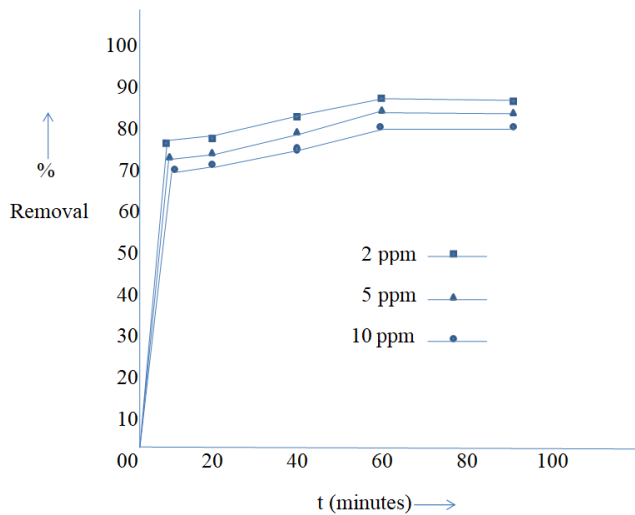


Figure 2: Effect of contact time on the removal of Cd (II) ions

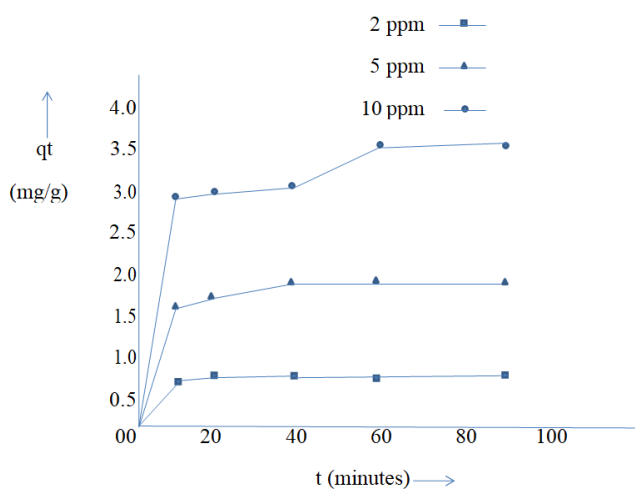


Figure 3: Effect of contact time on the adsorption of different concentrations of Cd (II) ions

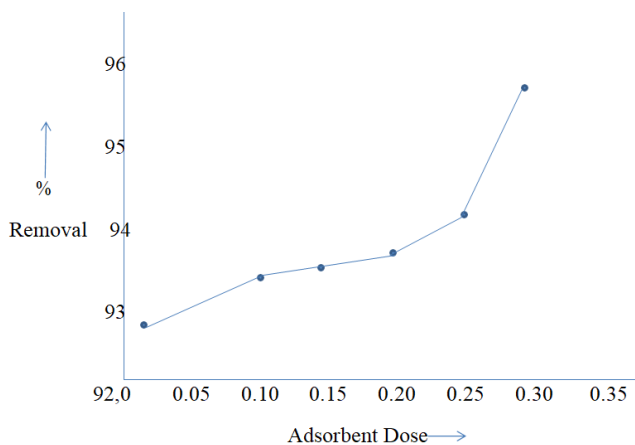


Figure 4: Effect of adsorbent dose on adsorption

3.3 Effects of adsorbent particle size:

Three different particle sizes of adsorbent (53, 150 and 350 μm) were investigated to study the effect of particle size on the adsorption of Cd (II). It was observed that adsorption capacity and removal efficiency increased with decreasing average particle size of adsorbent. When the particle size was increased from 53 μm to 350 μm , percentage removal decreased from 93% to 89% (Figure: 5).

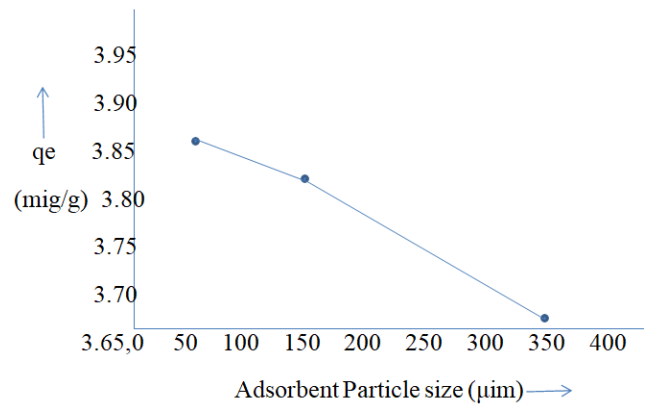


Figure 5: Effect of adsorbent particle size on the adsorption of Cd (II)

3.4 Adsorption isotherms:

Adsorption isotherm indicates relationship between the adsorbate in the fluid phase (solution) and the adsorbed on the adsorbent at constant temperature [30-31]. They are very useful for obtaining the adsorption capacity so as to facilitate the evaluation of the feasibility of the adsorption process for a given application and for selection of the most appropriate adsorbent at the optimum experimental conditions [30]. In this work, adsorption isotherms were carried out at initial concentrations of 2-20 mg/L. The Langmuir, Freundlich and Temkin isotherm models were employed to interpret the adsorption process in order to understand the mechanism of Cd (II) ion adsorption on Activated carbon. The data did not fit well into Temkin and Langmuir isotherms as their correlation coefficients are relatively lower than Freundlich model, which gave the best fit (Table:1).

Table 1: Isotherm model parameters for the adsorption of Cd (II)

Isotherm models	Parameters	Values
Langmuir model	R ²	0.915
	qm	7.810
	Ka	0.899
Freundlich model	R ²	0.995
	KF	3.370
	1/n	0.494
Temkin model	R ²	0.887
	β	1.291
	KT	23.23

The Langmuir isotherm equation [32] is given as,

$$q_e = \frac{q_m K_a C_e}{1 + K_a C_e} \quad (4)$$

Where, q_e is the amount of Cd (II) adsorbed per unit mass at equilibrium (mg/g), q_m is the maximum possible amount of Cd (II) that can be adsorbed per unit mass of adsorbent (mg/g), C_e is the concentration of adsorbate in the solution at equilibrium (mg/l) and K_a is adsorption equilibrium constant.

The linear form of above equation will be written as,

$$\frac{C_e}{q_e} = \frac{1}{K_a q_m} + \frac{C_e}{q_m} \quad (5)$$

A plot of C_e/q_e versus C_e gives a straight line, with a slope of $(1/q_m)$ and intercept $(1/K_a q_e)$. The essential characteristics of Langmuir isotherm can be expressed in terms of a dimensionless constant K_R , the separation factor or equilibrium parameter, which is defined as:

$$K_R = \frac{1}{1 + K_a C_0} \quad (6)$$

Where, K_R , K_a and C_0 are dimension less separation factor, Langmuir constant (L/mg), and initial concentration of Cd (II) ions (mg/L) respectively.

If $K_R = 1$ shape of the isotherm will be linear, If $K_R < 0$ it is irreversible,

If $K_R > 1$ unfavorable and If $0 < K_R < 1$ favorable.

(i) The Freundlich isotherm is an empirical model which indicates the surface heterogeneity of the adsorbent. The equation is given as:

$$q_e = K_F \cdot C_e^{1/n} \quad (7)$$

Taking logarithm both sides we get,

$$\log q_e = \log K_F + (1/n) \log C_e \quad (8)$$

Where, q_e = the amount of adsorbate adsorbed at equilibrium (mg/g); K_F (L/g) and n = Freundlich constants which indicate the adsorption capacity of the adsorbent and adsorption intensity, respectively; C_e = the

equilibrium concentration of adsorbate in the solution (mg/dm³).

A plot of $\log q_e$ vs $\log C_e$ gives a straight line of slope $1/n$ and intercept $\log K_F$ from which n and K_F can be evaluated.

If $1/n < 1$, then the adsorption is favorable and the adsorption capacity increases with the occurrence of new adsorption sites. But

if $1/n > 1$, then the adsorption bond becomes weak and unfavorable adsorption takes place, leading to a decrease in adsorption capacity.

(ii) Temkin studied the heat of adsorption and the adsorbent-adsorbate interaction of surfaces [33]. The Temkin isotherm is given as,

$$q_e = \left(\frac{RT}{b}\right) \ln K_T C_e \quad (9)$$

The linear form of equation is given as,

$$q_e = \left(\frac{RT}{b}\right) \ln K_T + \left(\frac{RT}{b}\right) \ln C_e$$

$$q_e = B_1 \ln K_T + B_1 \ln C_e \quad (10)$$

Where,

$$B_1 = \left(\frac{RT}{b}\right)$$

is related to the heat of adsorption and T = temperature in Kelvin. Temkin constants are obtained from the plot of q_e versus \ln

C_e . Such isotherm assumes chemical adsorption of adsorbate onto adsorbent.

3.5 Adsorption kinetics

The study of the adsorption kinetics of adsorption process is very important as it describes the rate of adsorbate uptake, which in turn evidently controls the residence time of the solute uptake at the solid- solution interface or the sorption reaction [33-35]. It is an important characteristic in defining the efficiency of sorption [33]. Generally, adsorption of adsorbate by an adsorbent consists of

several steps which include:

- Transport of adsorbate (solute) from the solution to the film surrounding the adsorbent particles. This is called bulk diffusion.
- Diffusion of the adsorbate from the film to the external surface. This is called external diffusion.
- Diffusion from the surface to the internal sites, known as intra-particle transport within the particle.
- Adsorption of the adsorbate on the interior surface of the sorbent known as pore diffusion.

In this study, the kinetic equations employed to investigate the mechanisms of cadmium ion adsorption are:

The Pseudo-first order equation is given as,

$$\frac{dq}{dt} = k_1 (q_e - q_t) \quad (11)$$

Table 2: Pseudo-first order Kinetics Parameters for the adsorption of Cd (II) by Activated carbon

Co (mg/L)	qe Exp	qe Cal	k ₁	R ²
2	0.779	0.029	0.040	0.927
5	1.888	0.380	0.039	0.976
10	3.488	1.294	0.049	0.949

The Pseudo-second order equation [36] is given as,

$$= k_2 (q_e - q_t)^2 \quad (14)$$

Where k₂ is the rate constant of pseudo-second order adsorption (g mg⁻¹ min⁻¹), The linear form of equation is given by,

$$\frac{t}{qt} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (15)$$

Table 3: Pseudo-second order Kinetics Parameters for the adsorption of Cd (II) by Activated carbon

Co (mg/L)	k ₂	qe Cal	k ₂ qe ²	R ²
2	3.33	0.781	2.031	1.0000
5	0.22	1.922	0.813	0.9998
10	0.07	3.602	0.908	0.9984

4. Conclusion

The present study was carried out to investigate the ability of activated carbon in the removal of cadmium ions from aqueous solution. The results showed that the equilibrium time for the adsorption of the ions onto adsorbent was 90 minutes and the adsorption process followed pseudo-second order kinetics. The data obtained fitted with Freundlich isotherm model (R²=0.9984). The monolayer adsorption capacity obtained from Langmuir isotherm was 7.81 mg/g. The adsorption is highly influenced by parameters such as contact time, adsorbent dosage and initial concentration of Cd (II). The study has shown that activated carbon has a considerable potential as an effective adsorbent for the removal of cadmium ions from aqueous solutions.

Where, q_e and q_t are the adsorption capacities at equilibrium and at time t (mg/g) respectively; k₁ is rate constant of pseudo-first order adsorption (min⁻¹).

After integration and applying boundary conditions t = 0 to t = t and q_t = 0 to q_t = q_e, equation (11) becomes,

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (12)$$

Where, q_e (mg g⁻¹) = the amount of Cd (II) adsorbed at equilibrium; q_t (mg g⁻¹) = the amount of Cd (II) adsorbed at time t and k₁ (min⁻¹) = the rate constant of pseudo-first order adsorption.

A plot of log (q_e - q_t) vs t gives the value of slope = k₁, and intercept = log q_e (Table 2).

A plot of t/q_t versus t gives the value of slope = q_e and intercept = k₂ (Table 2). From the result it is clear that pseudo-second order model give the best description of the mechanism of Cd (II) adsorption onto Activated carbon. Apart from its having the highest R² of 1, the adsorption capacity calculated from this model (0.781 mg/g) was very close to the experimental value (0.779 mg/g).

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