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# Removal of $Cd^{2+}$ from Aqueous Solution using Eucalyptus Sawdust as a Bio-Adsorbent: Kinetic and Equilibrium Studies

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## Abstract

Heavy metals are not dissoluble in the environment and can be dangerous for many species. So, removal of heavy metals from the water and wastewater is an important process. In this research, eucalyptus sawdust was prepared and employed for removal of Cadmium ions from aqueous solution. For this purpose, several parameters such as pH of aqueous solution, adsorbent dosage, contact time, initial concentration of cadmium ion and mixing rate were studied. The results showed that the best conditions for the removal of $Cd^{2+}$ were obtained at temperature of 30°C, mixing rate of 200 rpm, pH of 8, adsorbent dosage of 5 g/l, and initial concentration of cadmium of 200 ppm which the removal efficiency was obtained 89.3 %. In order to study the kinetics of adsorbent, the pseudo-first order and pseudo-second order kinetic models and intra-particle diffusion model were applied. According to the correlation coefficient ($R^2$), pseudo-second order kinetic model showed better correlation for kinetic behavior of the adsorbent. Furthermore, to study the equilibrium behavior of adsorbent, Langmuir and Freundlich models used and results showed that the Langmuir isotherm model had better matching with experimental data. So, this adsorbent can be used as natural and cheap adsorbent to remove $Cd^{2+}$ ions from aqueous solutions.

**Keywords:** Cadmium ions, Kinetic models, Isotherm models, Eucalyptus sawdust, Adsorption

## 1 Introduction

Heavy metals are not dissoluble in the environment and can be dangerous for many species [1, 2]. Heavy metals can also lead to changes in physical, chemical and biological properties of water [3-5]. Rapid improvements in economical industries like forgery, production of fertilizers, battery manufacturing and etc. are leading to direct and indirect increase in production rate of heavy metals into the environments of developing countries [6]. There are other industries which have heavy metals as byproducts including automotive industry, dyes for the textile industry and mining operations [7, 8]. As a result, recovery and removal of heavy metals from the water and the waste water is a significant process to maintain general and environmental health [1]. Cadmium is one of the most dangerous heavy metals and can be hazardous for human health causing serious diseases like kidney failures, hypertension, hepatitis and damage to the lungs and bones cancer [9]. The amount of cadmium in swage water and drinking water is reported to be equal to 0.1 and 0.05 mg/l, respectively [10].

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There are a number of different methods for recovery and removal of heavy metals in soil and aqueous solutions like electrochemical processes, ion exchange, chemical scaling, osmosis, evaporation and surface adsorption [11]. Some of these methods have disadvantages like high capital costs and/or inapplicability in industrial scale [12]. Therefore, some researches have focused on low cost and available adsorbent with agricultural and biological origins [13]. Some of these materials are sunflower wastes [14], orange peel [15], tea factory wastes [16], sawdust [17-18], soybean straw [19], olive stone [20, 21] and etc. These materials have been utilized by researchers for recovery and removal of heavy metals.

The main aim of this investigation is to assess the removal of cadmium ion from aqueous solutions using eucalyptus sawdust. To do so, the effect of several parameters such as pH, adsorbent dosage, initial concentration of cadmium ion in the solution, mixing rate and contact time on adsorption process were investigated. In addition, kinetic and equilibrium behavior of bio-adsorbent were examined.

## 2 Materials and Methods

### *2.1 Preparation of Cadmium solution*

In order to prepare cadmium aqueous solution, 2.744 g of cadmium nitrate $Cd(NO_3)_2.4H_2O$ is dissolved into one liter of distilled water. This solution is used for preparation of different concentrations of cadmium (3, 10, 15 and 20 ppm). In all experiments, diluted solution and distilled water were employed.

***2.2 Preparation of adsorbent***

Eucalyptus sawdust was gathered and washed with distilled water till no soil particles were inside the material and the rinsed water remained colorless. Thereafter, sawdust was put inside the oven and heated up to 70°C for 24 hours to remove humidity. The sawdust powder was prepared by household milling machine (Moulinex) and passed sieve No. 25 (ASTM E 11) and stored in poly-ethylene containers.

***2.3 Batch adsorption test and optimized conditions***

The adsorption experiment was carried out as batch inside 250 ml Erlenmeyer flasks containing 100ml of synthetic cadmium solution. Initial pH of the samples was set by 0.1 molar NaOH and HCl in the range of 2-10. Afterwards, 2g/l of adsorbent was added up to the solution with cadmium initial concentration of 10 ppm. The final solution was stirred at 30°C and 200 rpm for 80 minutes. The solutes were filtered through filter paper and 5ml of the solution was analyzed to measure adsorbed cadmium ion concentration. The optimization process repeated for other parameters as well as pH. These parameters were adsorbent dosage (1-8 g/l), mixing rate (0-200 rpm), and initial concentration of cadmium ion in the solution (3-20 ppm). The pH of the solution was adjusted on optimum condition and one of the parameter considered variable while others were constant. The optimized condition of each parameter was selected and investigation continued to define the optimum condition of other parameters. The cadmium ion concentration was identified by flame atomic adsorption spectroscopy model SpectrAA-10 plus made by Varian. The amount of adsorbed ions by bio-adsorbent for each gram of adsorbent is identified by Eq. (1) [22]:

$$q_e=(C_0-C_e)V/W \qquad (1)$$

in Eq. (1), qe is the amount of adsorbed material per gram of bio-adsorbent (mg/g) in equilibrium state, $C_0$ and $C_e$ are initial and equilibrium concentrations of cadmium (mg/l), V is the solution volume and W is the weight of adsorbent. In the present study, the recovery of lead ion adsorption in different conditions of the reaction is identified using Eq. (2) like below:

$$\%\ Adsorption=[(C_i-C_o)/C_i]*100 \qquad (2)$$

$C_i$ and $C_o$ are initial and final concentration of lead concentration, respectively in aqueous solution after equilibrium. In this research, all of the tests performed twice and the best results were reported.

***2.4 SEM Analysis***

The change on the surface of eucalyptus sawdust was studied before and after cadmium ion adsorption using SEM apparatus (Hitachi type, S4160). For scanning the surface of the adsorbent, these surfaces were covered with a thin layer of gold both before and after adsorption. The SEM images showed the apparent surface of the adsorbent. Figure 1 shows the surfaces of eucalyptus sawdust before and after cadmium ion adsorption.

## 3 Results and Discussions

***3.1 Effect of pH***

The capacity of ion metal adsorption and its mechanism depends on initial pH of the solution [23]. Adsorption recovery is greatly dependent on hydrogen cation in the solution [1]. The effect of initial pH performed in the range of pH = 2-10 and the percentage of removed cadmium ion is shown in Figure 2. This figure shows that increasing initial pH leads to increase in cadmium ion removal percentage and optimum pH is 8 and removal recovery is also 80%. Low pH means high hydrogen cation content and this cation participates in reactions inside the aqueous solution. In fact, there is a competition between hydrogen cation and cadmium ions to occupy active sites of the adsorbent. If these sites were occupied by hydrogen ions, there would be less active site for cadmium ions and this reduces the recovery of adsorption. High pH (pH > 7) means low hydrogen cation content and high $OH^-$ content inside the solution. In this case, there is no competition between hydrogen and cadmium ions and active sites are occupied by cadmium ions, resulting in higher recovery. In higher pH values (pH > 9) the adsorption recovery is reduced again. In this case, hydroxide anions compete with cadmium ions to lodge into active sites. Besides, in high pH values, hydroxide anions make a complex with cadmium ions and these ions deposit and accumulate in the solution. Cadmium removal percentage in pH = 10 is 77.5%.

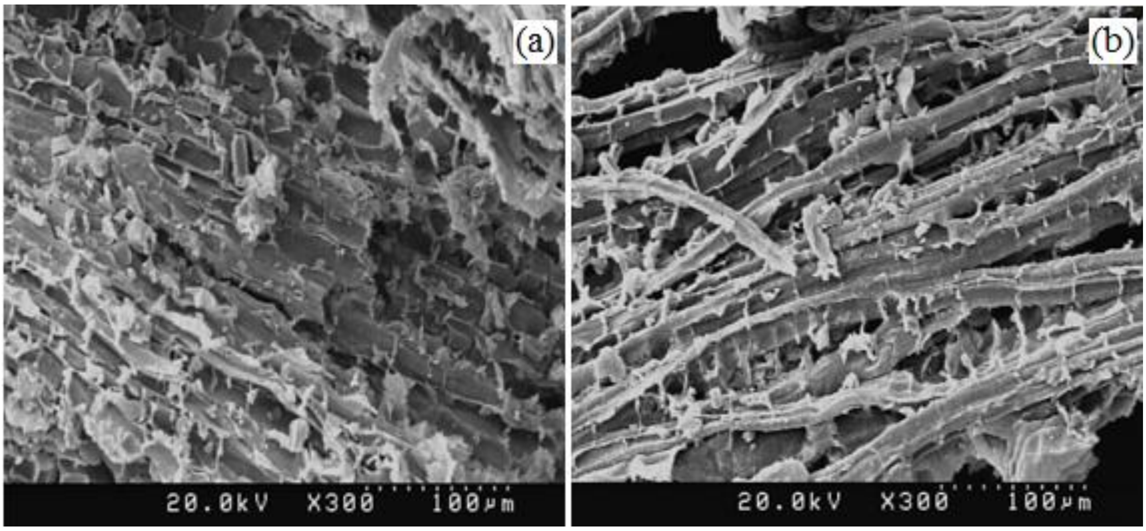



Figure 1: SEM images of the adsorbent a) eucalyptus sawdust before adsorption, b) sawdust after adsorption

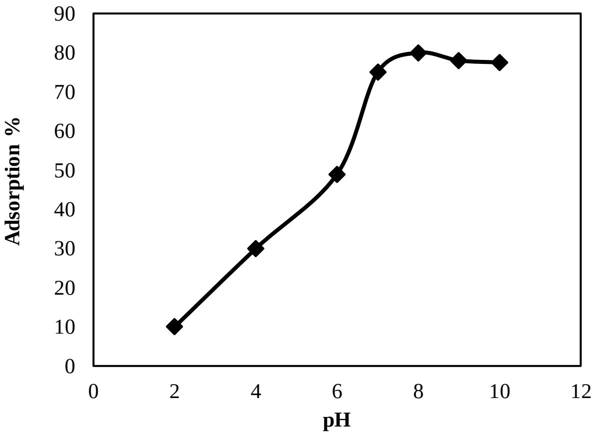



Figure 2: Effect of initial pH on efficiency of cadmium adsorption (initial cadmium concentration 10 ppm, revolution rate 200 rpm, temperature 30°C, contact time 80 minutes, adsorbent dosage 2 g/l)

### *3.2 Effect of mixing rate*

The mixing rate and turbulence making inside aqueous solution is an important parameter in adsorption process as disturbance increases the possibility of contact between the adsorbent and ions which results into higher recovery [24]. In order to examine mixing rate on adsorption process, the parameters were determined in laboratorial conditions as: mixing rate 0 -200 rpm, initial cadmium ion 10 ppm, time 80 minutes, temperature 30°C, adsorbent dosage 2 g/l and pH 8. Magnetic mixer was used for mixing process and this parameter effect is shown in Figure 3. According to Fig. 3, increasing the mixing rate in the range of 0-200 rpm, increases the recovery of cadmium adsorption by the adsorbent. Higher mixing rate means higher contact possibility between active sites and cadmium ions. The optimum recovery was in 200 rpm mixing rate equal to 80%.

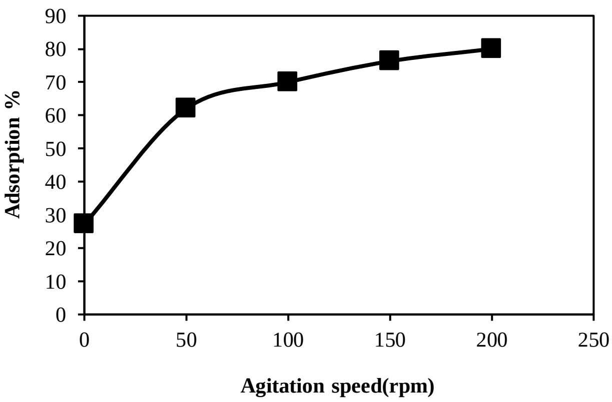



Figure 3: Effect of mixing rate on cadmium adsorption (initial cadmium concentration 10 ppm, temperature 30°C, contact time 80 min, adsorbent dosage 2 g/l, pH = 8)

### *3.3 Effect of adsorbent dosage*

The amount of used adsorbent is a significant parameter in adsorption process, as it determines the adsorption capacity in a certain concentration of adsorbed material [25, 17]. The testing parameters for the range of adsorbent dosage (1-8 g) to remove cadmium ions are: initial cadmium concentration 10 ppm, temperature 30°C, contact time 80 minutes, mixing rate 200 rpm, pH = 8. The findings revealed that increasing the amount of used adsorbent leads to increase in cadmium adsorption, as higher dosage increases the number of active sites for cadmium ion adsorption. Increase in the amount of adsorbent up to 5 g/l, results into higher recovery of cadmium removal that is 85%. Higher dosages of adsorbent in aqueous solution, more than 5 g/l, show negligible increase in recovery because of saturation in active sites. Even in some cases the final recovery is reduced as there was higher possibility of contact between adsorbent particles and active sites and this factor makes flocculation on the sites and ultimately decreases the number of active sites and adsorbent surfaces and final recovery as a result.

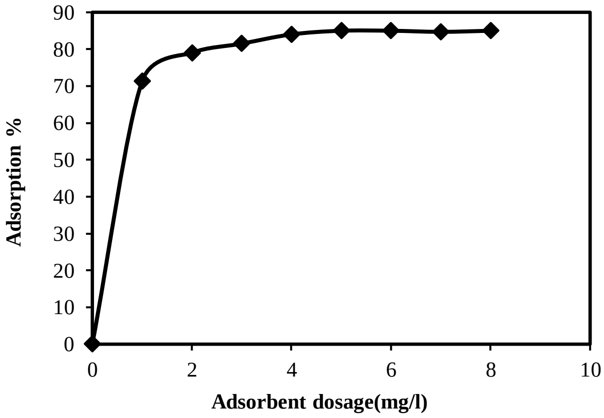



Figure 4: Effect of adsorbent dosage on cadmium adsorption (initial cadmium concentration 10 ppm, temperature 30°C, contact time 80 min, mixing rate 200 rpm, pH = 8)

### *3.4 Effect of initial concentration of cadmium ion and contact time*

In batch adsorption processes, initial concentration of metals plays an important role in providing appropriate force for transferring stable mass between solid and liquid phases [26]. The testing conditions of different initial concentration of lead ion (3-20 ppm) were: temperature 30°C, contact time 0-150 minutes, mixing rate 200 rpm, and pH = 8. Figure 5 shows the effect of initial cadmium ion (3, 10, 15, 20 ppm) in different contact times (0-150 min) for removal of $Cd^{2+}$ from synthetic wastewater using eucalyptus sawdust. This figure confirms that increasing both initial concentration of cadmium ion and contact time, amplifies the recovery; due to appropriate produced forces for mass transfer between solid and liquid phases. In addition, Figure 5 shows that longer contact time increases adsorption recovery. Cadmium ion adsorption rate by the adsorbent is higher during initial time intervals that because of activity of adsorption sites. Equilibrium time of adsorption by adsorbent is 20 min. After this period, the percentage of total adsorbed metal changes negligibly, since active sites were occupied by cadmium ions and adsorption continued through ions penetration into adsorbent layers. The results of this stage of the experiments were applied to investigate the kinetic behavior of prepared adsorbent.

### *3.5 Kinetic studies*

Adsorption kinetics is used for identification and control mechanisms of surface adsorption processes. This mechanism depends on physical and chemical properties of adsorbent. In the present study, kinetics and cadmium adsorption mechanism of the adsorbent were modeled by pseudo-first and pseudo-second order kinetic models and intra-particle diffusion model. The degree of this correlation was determined by correlation coefficient ($R^2$). The pseudo first order kinetic model assumes that the rate of changes in the solute is directly proportional to the changes in the saturation concentration and the amount of consumed adsorbent versus time. The linear form of the pseudo-first order model is as equation 3 [25, 27]:

$$\mathrm{Ln}\,(q_e - q_t) = \mathrm{Ln}\,q_e - K_1 t \tag{3}$$

In this equation, $q_e$ is the amount of adsorbed ion in equilibrium state (mg/g) per gram of the adsorbent, $q_t$ is the amount of adsorbed ion (mg/g) per gram of adsorbent versus time and k is the constant rate of adsorption (1/min).

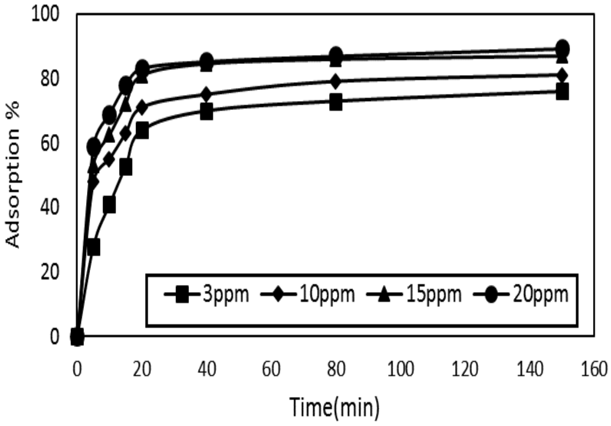



Figure 5: Effect of initial concentration of cadmium ion on adsorption using eucalyptus sawdust (temperature 30°C, mixing rate 200 rpm, pH = 8, adsorbent dosage 5 g/l)

This constant value is obtained by plotting Ln ($q_e$-$q_t$) versus t. Another kinetic model frequently used is pseudo-second order model and the linear form of this equation is like Eq. (4) [25]:

$$\frac{t}{q_t} = \frac{1}{k_2\, q_e{}^2} + \frac{t}{q_e} \tag{4}$$

In this equation, $K_2$ is the constant rate of pseudo-second order equation (g/mg.min), $q_e$ is the maximum adsorption capacity (mg/g) and $q_t$ is the adsorbed amount during the time t (min). Initial rate of adsorption is determined using Eq. (5) [25]:

$$H = K q_e^2 \tag{5}$$

The values of $q_e$ and $K_2$ are obtained by plotting $t/q_t$ versus t. $q_e$ is the slope of the linear plot and $K_2$ is the intercept of the line. Another kinetic model is intra-particle diffusion model which is as below:

$$q_t = K_{id} t^{1/2} + C \tag{6}$$

$q_t$ (mg/L) is the amount of adsorption time t (min) and $k_{id}$ (mg/g.min) is the rate constant of intra-particle diffusion. Table 1 list the parameters and constants of pseudo-first, pseudo-second and intra-particle diffusion models for eucalyptus sawdust in optimum conditions and for different concentrations of cadmium ion (3, 10, 15, 20 ppm) and the linear correlation between these parameters are shown in Figures 6 to 8.

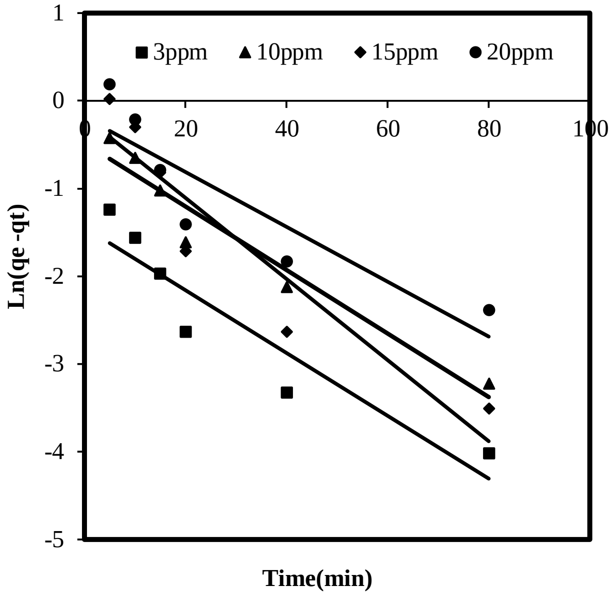
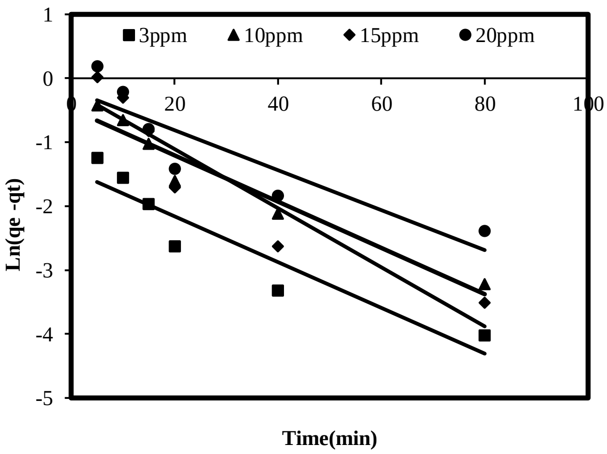



Figure 6: Kinetic diagram of pseudo-first order model using adsorbent prepared by eucalyptus sawdust (temperature 30°C, pH = 8, mixing rate 200 rpm, adsorbent dosage 5 g/l)



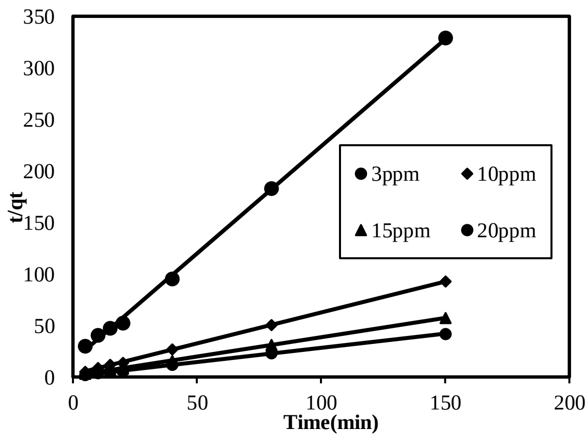




Figure 7: Kinetic diagram of pseudo-second order model using adsorbent prepared by eucalyptus sawdust (temperature 30°C, pH = 8, mixing rate 200 rpm, adsorbent dosage 5 g/l)

Based on the correlation coefficient for mentioned adsorbent, the pseudo-second order kinetic model is more appropriate in comparison with other models to describe the kinetic behavior of adsorbent and best matches with laboratorial data.

***3.6 Isotherm models***

The isotherm models are usually investigated for description of adsorption process and related mechanisms [28]. Langmuir and Freundlich are two isotherm models that are widely used. Langmuir isotherm is widely used for description of laboratorial data in previous studies. The linear form of this model is like below [25, 27-29]:

$$1/q_e=(1/k_L q_{max})1/C_e+1/q_{max} \quad (7)$$

$C_e$ is the concentration of metal ion in equilibrium state (mg/l), $q_e$ is the adsorbed ion in equilibrium state per gram of adsorbent. $q_{max}$ and $k_L$ are the capacity of surface adsorption (mg/g) and adsorption energy (l/g), respectively which are the constants of Langmuir model.

The values are obtained by calculating the slope and intercept of linear Langmuir equation in $C_e/q_e$ versus $C_e$ diagram. Another effective parameter in Langmuir equation is $R_L$ that describes the properties of the equation. The value of $R_L$ is the representative of the state and quality of adsorption isotherm model. If $R_L$ >1, $R_L$ =0, $R_L$ =1 and 0 <$R_L$< 1, the process is non-desired, irrevocable, linear and desirable, respectively [25, 29]. The value of $R_L$ is identified using Eq. (8):

$$R_L=1/(1+k_L C_0) \quad (8)$$

$C_0$ in Eq. (8) is the initial concentration of cadmium ion in aqueous solution in mg/l. Another typical isotherm model frequently used is Freundlich isotherm model. This model is empirical and able to describe adsorption of organic and inorganic compounds by different adsorbent. The non-linear Freundlich isotherm model is like Eq. (9):

$$q_e=K_f C_e^{1/n} \quad (9)$$

The linear form of this equation is like Eq. (10) which is used in this study:

$$Lnq_e= LnK_f + 1/n\ LnC_e \quad (10)$$

$q_e$ is the capacity of equilibrium adsorption (mg/g), $C_e$ is the equilibrium concentration of cadmium ion in the solution (mg/l), $K_f$ and n are the constants of Freunlich model that show the relationship between adsorption capacity and adsorption intensity, respectively. In order to identify these parameters ($K_f$ and 1/n), $Lnq_e$ is plotted versus $LnC_e$ and the slope of the line is 1/n and the intercept is $K_f$. The value of n in many researches is in the range of 1-10. High values of n represent the high interactions between the adsorbent and metal ions and n = 1 shows the linear adsorption for all active sites of the adsorbent [29, 30].

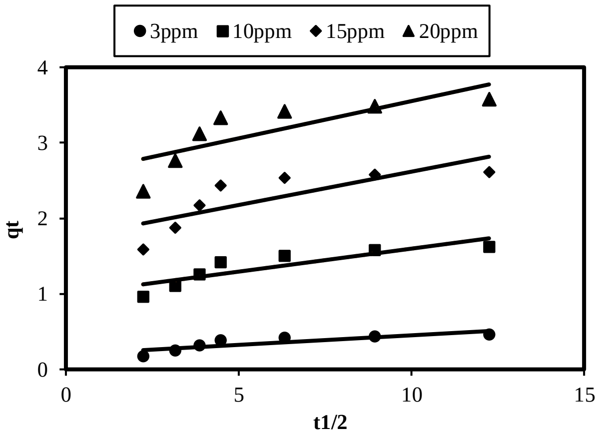


Figure 8: Kinetic diagram of intra-particle diffusion model using adsorbent prepared by eucalyptus sawdust (temperature 30°C, pH = 8 and 9, mixing rate 200 rpm, adsorbent dosage 5 g/l)

To investigate the equilibrium behavior of the process, test performed in the following conditions: initial cadmium ion concentration 10 ppm, temperature 30°C, contact time 80 minutes, mixing rate 200 rpm, adsorbent dosage 1-5 g/l and pH 8. Attained equilibrium data was examined for the adsorbent in Langmuir and Freundlich models. Figure 9 and 10 shows the equilibrium diagram of Freundlich and Langmuir for mentioned adsorbent. The constant values and other parameters of the models are listed in Table 2.

Table 1: Constants and correlation coefficient of kinetic models for cadmium ion adsorption in different concentrations using eucalyptus sawdust

| Kinetic models | Adsorbate concentration(mg/L) | | | |
|---|---|---|---|---|
| | 3 ppm | 10 ppm | 15 ppm | 20 ppm |
| Pseudo-first order | | | | |
| $q_{e\ cal}$ | 0.236 | 0.618 | 0.833 | 0.829 |
| $K_1$ | 0.0358 | 0.0362 | 0.0462 | 0.0313 |
| $q_{e.exp}$ | 0.456 | 1.62 | 2.61 | 3.572 |
| $R^2$ | 0.8763 | 0.9414 | 0.8778 | 0.7946 |
| Pseudo-second order | | | | |
| $q_{e.cal}$ | 0.481 | 1.666 | 2.671 | 3.627 |
| $K_2$ | 0.36 | 0.14 | 0.123 | 0.105 |
| $R^2$ | 0.999 | 0.9999 | 0.9998 | 0.9999 |
| H | 0.0833 | 0.388 | 0.877 | 1.381 |
| $q_{e.exp}$ | 0.456 | 1.62 | 2.61 | 3.572 |
| Intraparticle diffusion | | | | |
| $K_{in}$ | 0.0253 | 0.0609 | 0.0883 | 0.0985 |
| C | 0.1981 | 0.9899 | 1.7351 | 2.567 |
| $R^2$ | 0.7002 | 0.7565 | 0.6362 | 0.6396 |

The correlation coefficient for eucalyptus sawdust using Langmuir and Freundlich model was 0.9693 and 0.8679, respectively. This confirms that Langmuir model is a better estimator of isotherm behavior of eucalyptus sawdust in cadmium ion adsorption. It should be mentioned that the value of $R_L$ is 0.273 indicating desirable adsorption.

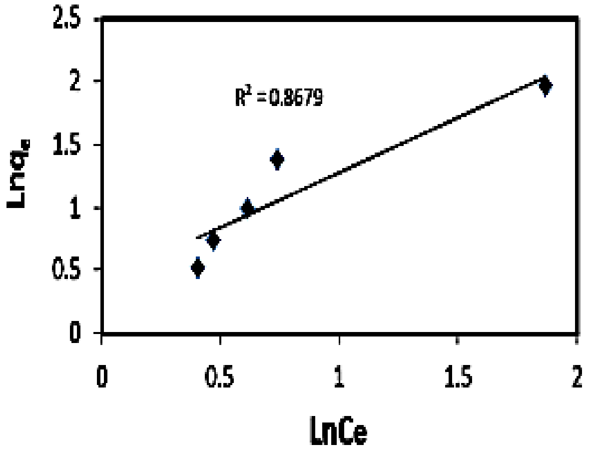



Figure 9: Freundlich adsorption isotherm plots for the adsorption of $Cd^{2+}$ ion

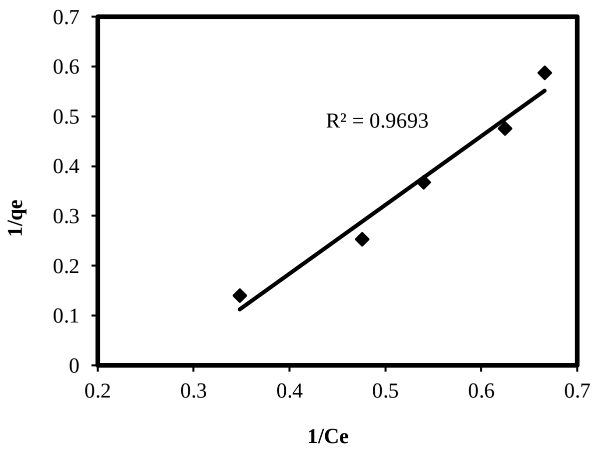



Figure 10: Langmuir adsorption isotherm plots for the adsorption of metal ion

Table 2: Constants and parameters of Langmuir and Freundlich isotherm models

| Adsorption isotherm | Isotherm constants | value |
|---|---|---|
| Langmuir | $K_L$(L/g) | 0.266 |
| | $q_{max}$(mg/g) | 2.716 |
| | $R_L$ | 0.273 |
| | $R^2$ | 0.9693 |
| Freundlich | $K_f$(mg/g)(L/mg)$^{1/n}$ | 1.495 |
| | n | 1.139 |
| | $R^2$ | 0.8679 |

## 4 Conclusion

The findings of this research were promoted for adsorption and removal of $Cd^{2+}$ ion from aqueous solution using bio-adsorbent prepared from eucalyptus sawdust. Effective parameters in cadmium ion adsorption were pH, the amount of used adsorbent, initial concentration of cadmium ion in the solution, contact time and mixing rate. The percentage of cadmium ion removal using eucalyptus sawdust was 89.3%. The optimum conditions were: temperature 30°C, mixing rate 200 rpm, adsorbent dosage 5 g/l, initial concentration of cadmium in the aqueous solution 20 ppm and pH of 8. Kinetic and isotherm behavior of the adsorbent were investigated by different synthetic and isotherm models and correlation coefficients of the adsorbent showed that pseudo-second order kinetic model was better estimators for kinetic behavior of adsorbent. Also, Langmuir isotherm model could well describe adsorptive behavior in comparison with Freundlich model. The value of $R_L$ for adsorbent was 0.273 indicating the adsorption process of cadmium ion by eucalyptus sawdust is desirable. Therefore, the recovery of ions removal and correlation coefficients of the models confirm that eucalyptus sawdust can be considered as natural and cheap adsorbent for cadmium ion removal.

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