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## Adsorption Studies of Coconut Shell Carbons Prepared by KOH Activation for Removal of Lead(II) From Aqueous Solutions

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**Abstract:** Removal of  $Pb^{2+}$  from aqueous solutions using coconut shell carbons produced by KOH activation is performed in this paper. Morphology and pore structure characteristic of coconut shell carbons are analyzed by SEM and nitrogen adsorption techniques. Effects of adsorbent concentration, agitation time and initial ion concentration on the adsorption behavior are investigated, and adsorption isotherm and kinetics on coconut shell carbons are also studied. The results show that high weight ratio of KOH/sample is favorable to produce rich porous structure. The resultant coconut shell carbons with a high specific surface area of  $1135\text{ m}^2/\text{g}$  is obtained and demonstrates good adsorption potential on removal of  $Pb^{2+}$  from aqueous solutions. Adsorption data fit well with Freundlich and Halsey isotherms. The kinetic studies indicate that adsorption behavior can be described by pseudo-second-order kinetic model, which also follows external diffusion and intra-particle diffusion in the adsorption process.

**Keywords:** coconut shell carbons; removal; porous; isotherm; kinetics

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### 1. Introduction

Water pollution caused by heavy metals has posed a significant threat to the environment and public health because of their toxicity, accumulation in the food chain and persistence in nature [1]. Among these heavy metals,  $Pb^{2+}$ , coming from battery manufacturing, ceramic and glass manufacturing,

metal plating and finishing, printing, and production of lead additives for gasoline, is known to have a severe toxic damage to neuronal system, kidneys, reproductive system, liver and brain [2,3]. Since lead does not degrade in environment like organic pollutants [4], the safe and effective disposal of wastewater containing  $Pb^{2+}$  is always a challenge to industrialists and environmentalists [5]. At present, various methods including chemical precipitation, electrochemical reduction, ion exchange, reverse osmosis, membrane separation, and adsorption have been developed to remove  $Pb^{2+}$  from wastewater [6–10]. Compared with other treatment method, adsorption appears to be an attractive process because it is simple, effective and economical in the removal of heavy metals from aqueous solution [11].

Activated carbons (ACs) exhibit a great adsorption capacity in wastewater and gas treatments as well as in catalysis, owing to their highly developed porosity, large surface area, and variable surface chemistry. However, high cost and non-renewable source of commercially available ACs limits its use as an adsorbent in developing countries [12,13]. In recent years, researchers have studied the production of ACs from cheap and renewable precursors, such as nutshells, fruit stones, coir pith, bagasse, bamboo, rice husk, and cotton stalks, *etc.* Coconut shell is a potential precursor for the production of ACs due to its excellent natural structure and low ash content. Conversion of coconut shells into activated carbons which can be used as adsorbents in water purification or treatment of industrial and municipal effluents would add value to these agricultural commodities, help reduce the cost of waste disposal, and provide a potentially cheap alternative to existing commercial carbons [14,15]. In this paper, systematic laboratory investigations of the removal of  $Pb^{2+}$  from aqueous solutions using coconut shell carbons as adsorbent have been reported and different models of isotherms and adsorption kinetics were fitted to the experimental data. The main objective of this research is to analyze adsorption behavior of  $Pb^{2+}$  on coconut shell carbons and evaluate its potential in removal of  $Pb^{2+}$  from the aqueous solution.

## 2. Experimental

### 2.1. Preparation and Characterizations of Coconut Shell Carbons

The coconut shells were cleaned with deionized water and dried at 110 °C for 48 h to reduce the moisture content. The dried samples were then crushed and sieved to a size range of 1–2 mm. Subsequently, coconut shells were carbonized in  $N_2$  gas up to the temperature of 500 °C at the rate of 20 °C/min and held for 2 h. After carbonization, these samples were mixed with water and KOH in a stainless steel beaker with the weight ratio of KOH/sample equal to 1:2 (CSC-A) and 2:1(CSC-B). Water was evaporated at 130 °C for 4 h, and these dried mixtures were heated in  $N_2$  gas at a rate of 10 °C/min to 800 °C, and kept at this temperature for 1 h. The products were cooled to room temperature and washed with HCl and deionized water until the pH of the washing solution reached 6–7.

The porous structure of coconut shell carbons was characterized by nitrogen sorption technique (Quantachrome Autosorb-iQ). The specific surface areas ( $S_{BET}$ ) of coconut shell carbons were analyzed by following Brunauer-Emmett-Teller (BET) method [16], and the total pore volume ( $V_T$ ) was calculated from the liquid volume of nitrogen at a relative pressure of 0.99. The morphology of coconut shell carbon was observed by a scanning electron microscope (SEM) (Philips XL30 FEG).

## 2.2. Adsorption Experiments

Simulated wastewater with different  $Pb^{2+}$  concentrations (100, 200, 300, 400, 600, 900 and 1200 mg/L) were prepared by dilution of the stock  $PbSO_4$  solution. Coconut shell carbons were added to 250 mL of  $Pb^{2+}$  solutions. After the adsorption processes, the samples were filtered through a 0.45  $\mu m$  membrane, and the filtrates were immediately analyzed by conductometer according to dependent relationship between water conductivity and  $Pb^{2+}$  concentration [17].

The sorption capacity  $q_e$  (mg/g) and removal efficiency  $Q$  were obtained according to the Equations (1) and (2), respectively:

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

$$Q = \frac{(C_0 - C_e)}{C_0} \times 100\% \quad (2)$$

where  $V$  is the volume of the solution,  $W$  is the amount of adsorbent,  $C_0$  and  $C_e$  are the initial and equilibrium concentration in the solution.

## 2.3. Adsorption Isotherm

$Pb^{2+}$  adsorption by coconut shell carbons were analyzed using Langmuir, Freundlich, Temkin, Dubinin–Radushkevich, Harkins–Jura and Halsey isotherms. The Langmuir isotherm is used to characterize the monolayer adsorption, which is represented by the following linear form:

$$\frac{C_e}{q_e} = \frac{1}{q_{max}b} + \frac{C_e}{q_{max}} \quad (3)$$

The essential characteristics of the Langmuir isotherm is expressed in terms of a dimensionless constant separation factor,  $R_L$ , which is defined as:

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

where  $q_e$  is the equilibrium adsorption uptake of heavy metal ions,  $q_{max}$  is the maximum adsorption capacity corresponding to the complete monolayer coverage.  $b$  is the Langmuir constant which is related to the energy of adsorption.  $C_m$  is the highest initial heavy metal ions concentration.

The Freundlich isotherm is generally applicable to the adsorption occurred on heterogeneous surface. The linear form is shown:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (5)$$

where  $K_F$  and  $n$  are Freundlich constants related to adsorption capacity and adsorption intensity, respectively.

Dubinin-Radushkevich (D-R) isotherm is expressed as:

$$\ln q_e = \ln q_m - \beta \varepsilon^2 \quad (6)$$

$$\varepsilon = RT \ln \left( 1 + \frac{1}{C_e} \right) \quad (7)$$

$$E = \frac{1}{\sqrt{2\beta}} \quad (8)$$

where  $\varepsilon$  is Polanyi potential,  $\beta$  is the Dubinin-Radushkevich constant,  $R$  is the gas constant (8.31 J/mol·K),  $T$  is the absolute temperature,  $E$  is the mean adsorption energy.

The Tempkin isotherm has been used in the following form:

$$q_e = B_T \ln K_T + B_T \ln C_e \quad (9)$$

$$B_T = \frac{RT}{b_T} \quad (10)$$

where  $K_T$  is Temkin adsorption potential,  $B_T$  is related to the heat of adsorption,  $b_T$  is the variation of adsorption energy.

Halsey isotherm is used to evaluate the multilayer adsorption at a relatively large distance from the surface, which is expressed by:

$$\log q_e = \frac{1}{n_H} \log K_H - \frac{1}{n_H} \log C_e \quad (11)$$

where  $K_H$  and  $n_H$  are the Halsey constants.

The Harkin-Jura isotherm is expressed as:

$$\frac{1}{q_e^2} = \frac{B_{HJ}}{A_{HJ}} - \frac{1}{A_{HJ}} \log C_e \quad (12)$$

where  $A_{HJ}$  and  $B_{HJ}$  are the Harkins-Jura constants.

#### 2.4. Adsorption Kinetics

In order to investigate the mechanism of adsorption, kinetic models such as the pseudo-first order, the pseudo-second order, and the intra-particle diffusion model were applied to study the adsorption dynamics.

The pseudo-first-order kinetic model can be expressed in linear form:

$$\log(q_e - q_t) = \log q_e - \frac{K_1 t}{2.303} \quad (13)$$

The pseudo-second-order kinetic model is used in the following linear form:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (14)$$

where  $k_1$  and  $k_2$  are the adsorption rate constants of pseudo-first-order and pseudo-second-order kinetic models, respectively,  $q_t$  is adsorption uptake at time  $t$ .

Spahn and Schlünder model is chosen to describe the external diffusion on the adsorbent:

$$\ln \frac{C_t}{C_0} = -k_{ext} t \quad (15)$$

where  $K_{ext}$  is external diffusion coefficient,  $C_t$  is concentration at time  $t$ .

The intra-particle diffusion model is expressed by:

$$q_t = k_p t^{1/2} + C \quad (16)$$

where  $k_p$  is the intra-particle diffusion rate constant,  $C$  is a constant related to the thickness of the boundary layer.

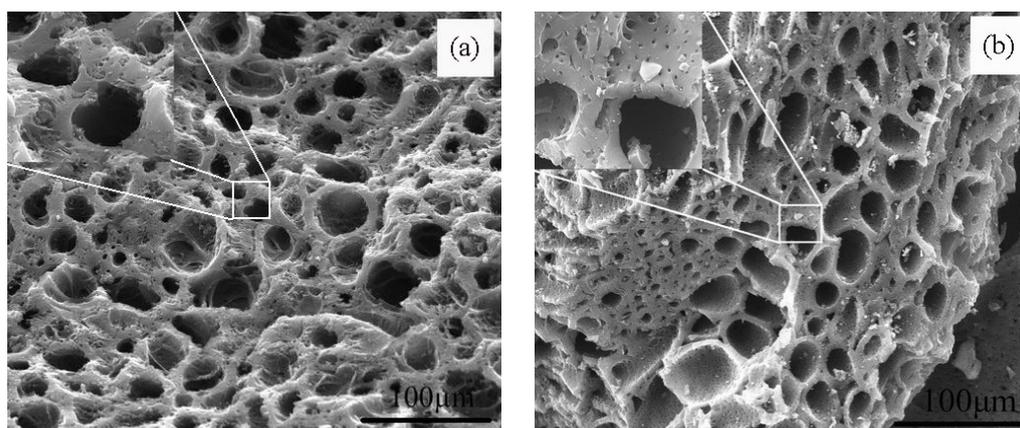
### 3. Results and Discussion

#### 3.1. Morphology and Pore Structure Properties of Coconut Shell Carbons

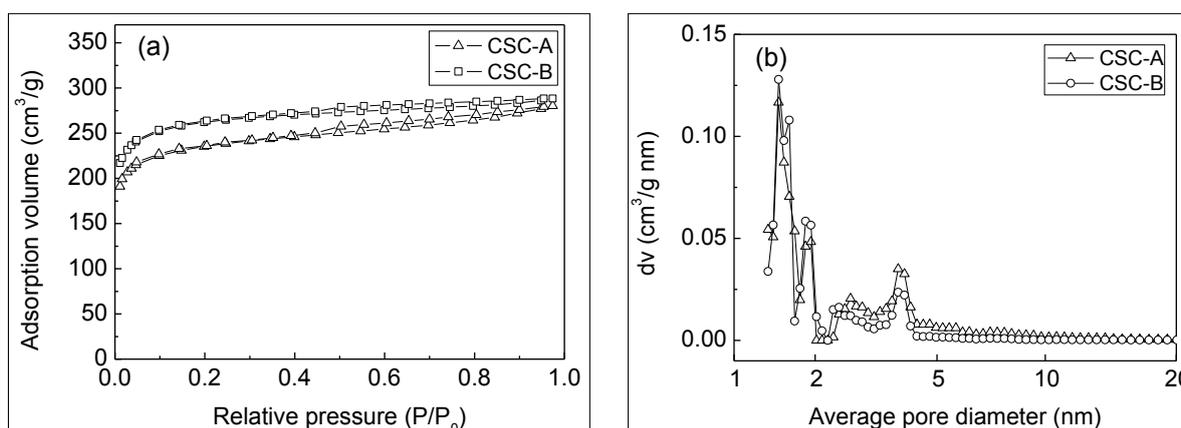
Typical SEM images of CSC-A and CSC-B are shown in Figure 1. Many large pores with honeycomb shape are formed on the surface of coconut shell carbons, which shows KOH is effective in creating well-developed pores in coconut shell carbons. Moreover, the high ratio of KOH/sample is favorable to produce more rich porous structure.

Figure 2 shows the typical adsorption/desorption isotherms of N<sub>2</sub> at 77 K for CSC-A and CSC-B. They both exhibit the typical type I isotherm according to the IUPAC classification, which suggests a predominantly microporous structure. However, there are significant differences in the N<sub>2</sub> adsorption volumes and the exact shape of the isotherms, depending on the weight ratio of KOH/sample. The isotherm of CSC-B exhibits a high volume of nitrogen adsorption and a narrow pore size distribution, which appears to imply that relative high ratio of KOH/sample is conducive for the development of microporosity [18].  $S_{\text{BET}}$  and  $V_{\text{T}}$  are directly related to the development of porosity of activated carbons. The increase in the ratio of KOH/sample from 0.5 to 2 promotes an improvement of the  $S_{\text{BET}}$  (from 728 m<sup>2</sup>/g to 1135 m<sup>2</sup>/g) and  $V_{\text{T}}$  (from 0.390 cm<sup>3</sup>/g to 0.442 cm<sup>3</sup>/g), producing more rich porous structure in coconut shell carbons, which is consistent with SEM analysis (Figure 1).

**Figure 1.** SEM micrographs of CSC-A (a) and CSC-B (b).



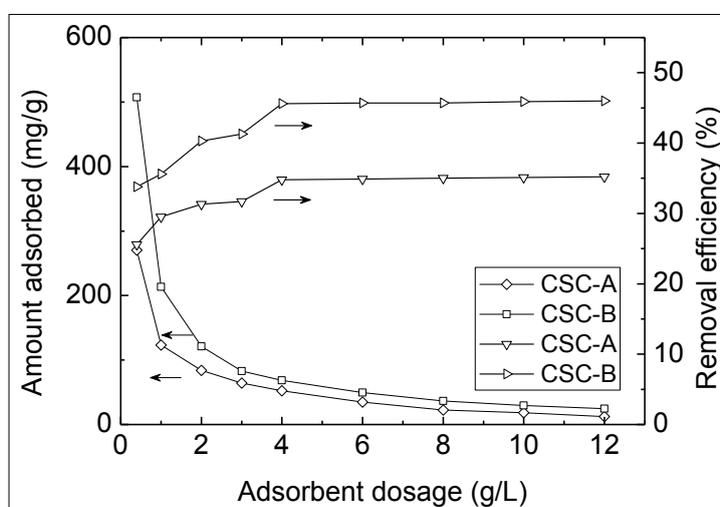
**Figure 2.** (a) Nitrogen adsorption–desorption isotherms and (b) pore size distributions for coconut shell carbons.



### 3.2. Effect of Adsorbent Concentration on $Pb^{2+}$ Removal

Figure 3 shows  $Pb^{2+}$  removal efficiency and adsorption capacity for CSC-A and CSC-B. It is clear that  $Pb^{2+}$  removal efficiency increases with increasing adsorbent concentration. This may be due to more active adsorption sites for  $Pb^{2+}$  at higher adsorbent concentration [19]. After adsorbent concentration reaches 4 g/L, no obvious increase is observed, which suggests the equilibrium between ions bound to the adsorbent and free ions is established [20]. Figure 3 also demonstrates that  $Pb^{2+}$  adsorption capacity decreases as adsorbent concentration increases. This reveals that more active sites are utilized at lower adsorbent concentration, producing a higher adsorption capacity, while only part of active sites are occupied by  $Pb^{2+}$  at higher adsorbent concentration, leading to a lower adsorption capacity [21]. We also note that adsorption of  $Pb^{2+}$  in aqueous solutions is related to  $S_{BET}$  and  $V_T$  of adsorbent [22]. CSC-B with larger  $S_{BET}$  and  $V_T$  had higher  $Pb^{2+}$  removal efficiency and adsorption capacity than CSC-A, suggesting its high affinity toward  $Pb^{2+}$  adsorption.

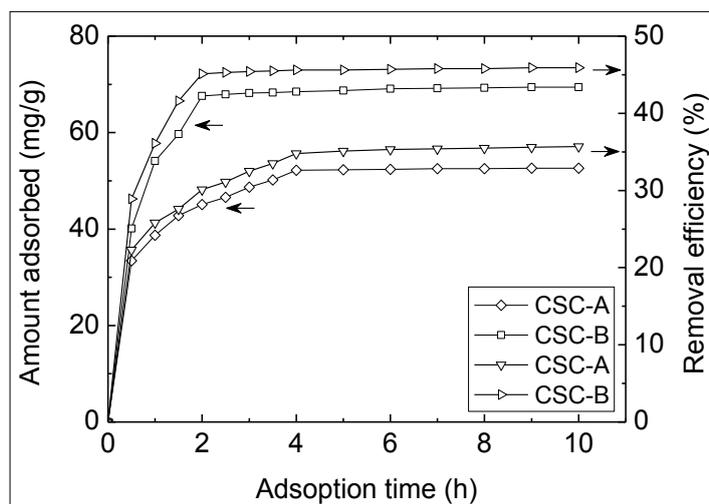
**Figure 3.** Effect of adsorbent concentration on amount adsorbed and removal efficiencies of coconut shell carbons.



### 3.3. Effect of Agitation Time on $Pb^{2+}$ Removal

Figure 4 presents the influence of agitation time on  $Pb^{2+}$  removal. Removal efficiency and adsorption capacity of CSC-A and CSC-B increase sharply in the initial stage and then gradually remain steady with the increase of agitation time. The initial fast adsorption may be attributed to large uncovered surface area of coconut shell carbons. With further increasing agitation time, the availability of the uncovered surface area gradually diminishes, and adsorption equilibrium is established [21,23]. Moreover,  $S_{BET}$  and  $V_T$  of the adsorbent have great influence on the time of adsorption equilibrium. It takes about 2 h to reach adsorption equilibrium for CSC-B, while about 4h for CSC-A, which can be explained due to the fact that high binding sites derived from high surface area shorten the time to reach equilibrium [22].

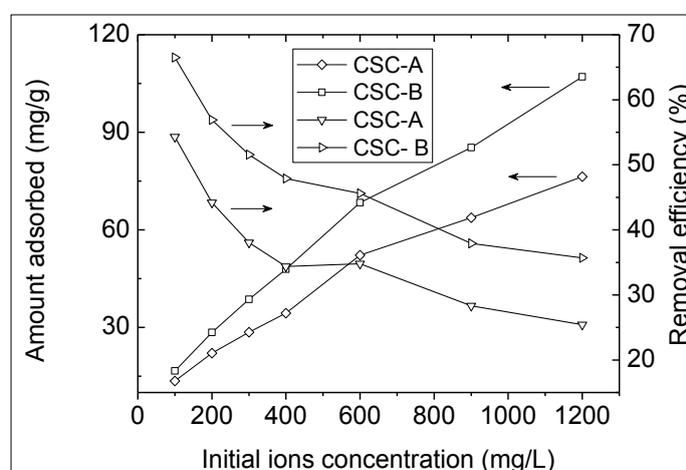
**Figure 4.** Effect of adsorption time on amount adsorbed and removal efficiencies of coconut shell carbons.



### 3.4. Effect of Initial Ions Concentration on $Pb^{2+}$ Removal

The affect of initial ions concentration on  $Pb^{2+}$  removal efficiency and adsorption capacity is shown in Figure 5. With the increase of initial ions concentration, the removal efficiency of CSC-A and CSC-B decrease from 54.3% and 66.5% to 25.4% and 35.7%, while adsorption capacity of CSC-A and CSC-B increase from 13.57 mg/g and 16.2 mg/g to 76.34 mg/g and 107.7 mg/g. This may be attributed to that sufficient adsorption sites are available at lower concentration, which facilitates the  $Pb^{2+}$  interaction with adsorption sites. However, in the case of higher concentration, adsorption sites of coconut shell carbons are saturated, leading to the decrease in the adsorption efficiency [24,25].

**Figure 5.** Effect of initial ions concentration on amount adsorbed and removal efficiencies of coconut shell carbons.



### 3.5. Adsorption Isotherm

The adsorption isotherms for  $Pb^{2+}$  removal were studied using initial concentration of  $Pb^{2+}$  between 100 mg/L and 1200 mg/L at an adsorbent dosage level of 4 g/L. Six adsorption isotherms (Langmuir,

Freundlich, Dubinin-Radushkevich, Tempkin, Halsey and Harkin-Jura isotherms) were adopted to investigate  $Pb^{2+}$  adsorption behavior on CSC-A and CSC-B. The parameters of the six adsorption isotherms are listed in Table 1.

**Table 1.** Parameters of six adsorption isotherms for  $Pb^{2+}$  adsorption on coconut shell carbons.

Parameter	CSC-A	CSC-B
<b>Langmuir isotherm</b>		
$q_{max}$ (mg/g)	112.36	151.52
$b$ (L/mg)	0.0021	0.0026
$R_L$	0.2815	0.2401
$R^2$	0.9288	0.9461
<b>Freundlich isotherm</b>		
$n$	1.6784	1.6753
$K_F$	1.3421	2.0188
$R^2$	0.9906	0.9972
<b>Dubinin-Radushkevich (D-R) isotherm</b>		
$q_m$	53.11	73.35
$\beta$	$9 \times 10^{-8}$	$7 \times 10^{-8}$
$E$ (kJ/mol)	2.36	2.67
$R^2$	0.7820	0.8162
<b>Tempkin isotherm</b>		
$b_T$ (kJ/mol)	23.82	17.92
$K_T$	5.77	7.38
$R^2$	0.9225	0.9270
<b>Halsey isotherm</b>		
$n_H$	-1.6784	-1.6753
$K_H$	0.6102	0.3082
$R^2$	0.9906	0.9972
<b>Harkin-Jura isotherm</b>		
$A_{HJ}$	263.15	416.67
$B_{HJ}$	2.79	2.63
$R^2$	0.8261	0.7891

A preliminary screening of the corresponding data has shown that Freundlich and Halsey isotherms fit the experimental data well due to high correlation coefficient ( $R^2$ ), which may be attributed to the heterogeneous distribution of active sites and multilayer adsorption on coconut shell carbons [26,27].  $K_F$  (2.0188) of CSC-B calculated from Freundlich isotherms is larger than  $K_F$  (1.3421) of CSC-A, indicating that CSC-B has high affinity toward  $Pb^{2+}$  because of high  $S_{BET}$  and  $V_T$ .  $R_L$  values of CSC-A and CSC-B obtained from Langmuir isotherm lie within the favorable limit between 0 and 1, revealing favorable adsorption of  $Pb^{2+}$  on coconut shell carbons. In addition, CSC-B has higher adsorption capacity  $q_{max}$  (151.52 mg/g) than CSC-A (112.36 mg/g), which also demonstrates that CSC-B has high adsorption ability toward  $Pb^{2+}$ . For Temkin isotherm, it is found that the  $R^2$  values of CSC-A and CSC-B are close to the values of Langmuir isotherm. The Temkin adsorption potential ( $K_T$ ) of CSC-B is larger than that of CSC-A, indicating a high adsorbent- $Pb^{2+}$  adsorption potential [28]. The mean adsorption energy calculated from D-R isotherm is 2.36 and 2.67 kJ/mol for CSC-A and CSC-B,

respectively, which implies the adsorption process can be considered as the physical adsorption. Harkins–Jura isotherm suggests the multilayer adsorption as well as heterogeneous pore distribution in the adsorbents surface. However, D-R and Harkin-Jura isotherm exhibit low  $R^2$  values, indicating that the adsorption process less follows the two models.

In order to assess the performance of coconut shell carbons as adsorbent for  $Pb^{2+}$  removal, a comparison with other types of adsorbents reported in the literature is carried out (Table 2). The adsorption capacity of CSC-A and CSC-B is roughly 112.36 mg/g and 151.52 mg/g, respectively [21], demonstrating great predominance respect to other adsorbents, which suggests that they are promising adsorbents to remove heavy metals from aqueous solutions.

**Table 2.** A comparison of the adsorption capacity of coconut shell carbons with the literature data.

Adsorbents	pH	Dosage (g/L)	$S_{BET}$ ( $m^2/g$ )	Adsorption capacity (mg/g)	Reference
Enteromorpha prolifera	5	0.5	1688	146.85	[8]
Pine cone activated carbon	5	2	1094.1	27.53	[29]
Palm shell activated carbon	5	5	957.04	95.2	[30]
Apricot stone	5	2	566	22.84	[31]
CSC-A	5	4	728	112.36	In this study
CSC-B	5	4	1135	151.52	In this study

### 3.6. Adsorption Kinetics

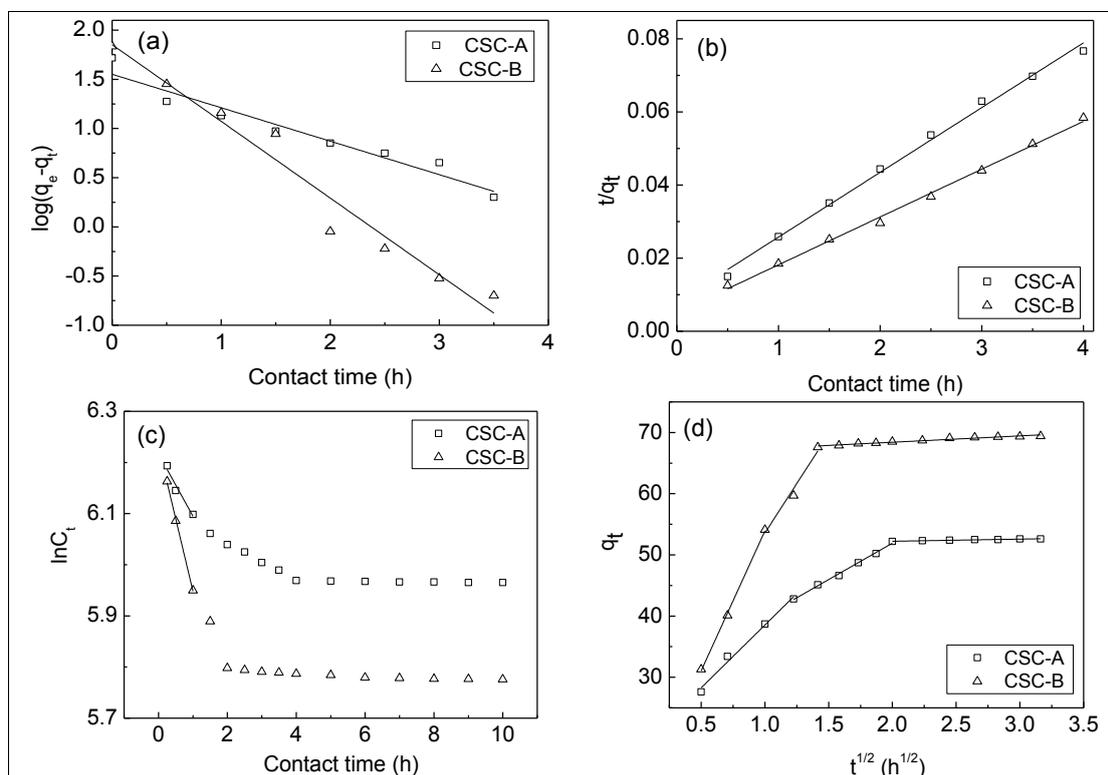
Adsorption kinetics is an effective method to evaluate the mechanism of  $Pb^{2+}$  adsorption on coconut shell carbons. Here, four adsorption kinetics models (pseudo-first-order model, pseudo-second-order model, Spahn and Schlünder model, and the intra-particle diffusion model) are applied to analyze the experimental data. As shown in Table 3 and Figure 6a,b, the adsorption data of CSC-A and CSC-B fit the pseudo second-order model perfectly judging by high correlation coefficients ( $R^2$ ), which suggests that  $Pb^{2+}$  adsorption on coconut shell carbons appeared to be controlled by a chemisorption process.

Figure 6c displays the plots of  $\ln C_t$  versus  $t$ . It is obvious that they follow a linear relationship in the initial stage of adsorption ( $t < 0.5$  h), which indicates that external diffusion is the rate-controlling step during this stage because of fast adsorption. Figure 6d and Table 3 show intra-particle diffusion model for  $Pb^{2+}$  adsorption on CSC-A and CSC-B, which present multi-linearity characterizations, indicating that three steps occurred in the adsorption process. The first sharper section is attributed to  $Pb^{2+}$  diffusion through the solution to external surface of adsorbent, so-called external diffusion. The second section describes the gradual adsorption stage, corresponding to  $Pb^{2+}$  diffusion inside the pores of the adsorbent, where intra-particle diffusion is rate-controlled. The third section is attributed to the final equilibrium stage [17,32].

**Table 3.** Parameters of pseudo-first-order, pseudo-second-order model, Spahn and Schlünder model, and intra-particle diffusion model for Pb<sup>2+</sup> adsorption on coconut shell carbons.

Parameter	CSC-A	CSC-B
<b>Pseudo-first-order kinetic model</b>		
$K_1$	0.7835	1.7961
$R^2$	0.9352	0.9578
<b>Pseudo-second-order kinetic model</b>		
$K_2$	0.0392	0.0336
$R^2$	0.9949	0.9962
<b>Spahn and Schlünder model</b>		
$K_{ext}$	0.1219	0.2827
$R^2$	0.9192	0.9981
<b>Intra-particle diffusion model</b>		
<b>Stage 1</b>		
$K_{i1}$	20.5391	45.7459
$R^2$	0.9842	0.9979
<b>Stage 2</b>		
$K_{i2}$	11.8941	32.3560
$R^2$	0.9946	0.9570
<b>Stage 3</b>		
$K_{i3}$	0.3556	1.0351
$R^2$	0.9545	0.9488

**Figure 6.** (a) pseudo-first-order model, (b) pseudo-second-order model, (c) Spahn and Schlünder model and (d) intra-particle diffusion model for Pb<sup>2+</sup> adsorption on coconut shell carbons.



#### 4. Conclusions

The coconut shell carbons prepared by KOH activation exhibit great advantage in  $Pb^{2+}$  removal from aqueous solution. High weight ratio of KOH/sample is favorable to produce coconut shell carbons with a high specific surface area and demonstrate good adsorption ability. The removal efficiency and adsorption capacity are great dependent on adsorbent concentration, agitation time and initial ion concentration. Adsorption data of coconut shell carbons can be represented by Freundlich and Halsey isotherms. Adsorption Kinetics obeys a pseudo second-order kinetic model and also follows external diffusion and intra-particle diffusion in the adsorption process.

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#### Conflicts of Interest

The authors declare no conflict of interest.

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