



## Uptake of hexavalent uranium from aqueous solutions using coconut husk activated carbon

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### ABSTRACT

Coconut husk activated carbon (CHAC) was used as an adsorbent to remove hexavalent uranium (U(VI)) from aqueous solutions by adsorption. Batch experiments were conducted to investigate the effects of contact time, pH, initial U(VI) concentration, and adsorbent dosage on U(VI) adsorption. The adsorption process reached an equilibrium state after 150 min. The uptake of U(VI) was highly pH dependent, and the optimum adsorption was at near-neutral pH. High U(VI) removal efficiencies could be achieved by increasing CHAC dosages. The uranium adsorption capacity increased with increasing initial U(VI) concentrations. Above a certain point, any further increase in initial U(VI) concentration produced no significant change in uranium adsorption capacity. Langmuir and Freundlich models were employed to describe the adsorption isotherm of U(VI) by CHAC. The Langmuir model fitted the equilibrium data better than the Freundlich isotherm. The maximum U(VI) adsorption capacity of CHAC was 6.67 mg/g. Kinetics data were also examined in terms of adsorption kinetics using pseudo-first-order and pseudo-second-order kinetic models. The results suggested that the adsorption process followed the pseudo-second-order kinetics well. In summary, CHAC had significant potential for the removal of U(VI) from aqueous solutions.

*Keywords:* Uranium; Adsorption; Coconut husk activated carbon; Isotherm; Kinetics

### 1. Introduction

Although nuclear power is an economical, safe, and clean form of energy for alleviating the global energy crisis, uranium mining and processing have led to the production of considerable amounts of

radioactive wastes, which cause long-term environmental effects. Uranium is one of the most dangerous heavy metals because of its high chemical toxicity and radioactivity. Once released into soil and water bodies, uranium can pose potentially serious health hazards to humans. Radioactive toxins can cause lung, kidney, and liver damage, cancer, leukemia and genetic aberrations [1,2]. The World Health Organization identifies

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uranium as a human carcinogen and sets a guideline limit of 15 µg/L of uranium in drinking water [3]. Therefore, the removal and recovery of uranium from wastewater is important because of the dual significance of uranium as a potential environmental threat and a nonrenewable energy source.

Conventional approaches for treating uranium-bearing effluents involve precipitation, reduction, electrochemical treatment, reverse osmosis, membrane filtration, solvent extraction, and ion exchange adsorption. These methods are often expensive and ineffective, particularly at low metal levels, thereby limiting their practical applications [4]. Hence, the development of new methods and materials for environmental remediation poses a serious challenge to environmental researchers. Adsorption has been proposed as a promising method to remediate uranium-containing wastewater because of its simple operation, mature technology, and extensive applicability.

Previous studies have suggested that bacteria, fungi, algae, and inorganic materials have certain uranium adsorption ability; however, their treatment effects are still unsatisfactory [5–10]. Numerous approaches have been investigated to develop cheap and effective uranium adsorbents. Considerable attention has been paid to agricultural and forestry by-products such as chaff, straw, corncob, plant leaves, and nutshell [11–15] because of their low cost, low secondary pollution, and easy recycling. Feng and Yi [16] investigated the adsorption of U(VI) on rice husk. They reported a maximum sorption capacity of 5.005 mg/g for U(VI) ions. Cao et al. [17] reported the rapid uptake of uranium from solution by chitosan and its derivants with a maximum sorption capacity of 2.5 mg/g. Wang et al. [18], working with banyan leaves, found that this biomass had a maximum removal capacity of 6.65 mg/g.

Coconut husk as an agricultural by-product is classified as a hard wood that contains cellulose, hemicellulose, lignin, and some other extractive compounds. Coconut husk is extensively used to develop various products such as shell handicrafts, shell powders, bear glasses, show pieces, and shell buttons. Coconut husk can also be processed into coconut husk activated carbon (CHAC) by high-temperature steam activation and refining. CHAC has microporous and mesoporous structures with large internal surface areas, thus enabling the use of CHAC as a high-performance adsorption material. To our knowledge, little research has been conducted on the adsorption of hexavalent uranium (U(VI)) by CHAC, although other wood activated carbon materials have been used in the removal of radionuclide. In this study, CHAC was applied to adsorb uranyl cations in the solution. The optimum

adsorption conditions were determined as a function of contact time, pH, initial U(VI) concentration, and adsorbent dosage. The adsorption isotherms and kinetics were also identified. The aim of this study was to evaluate the feasibility of CHAC as a sorbent to eliminate U(VI) contamination.

## 2. Materials and methods

### 2.1. Adsorbent, chemical reagents, and U(VI) stock solution

CHAC powder was purchased from Guoqing Water Purification Material Co. Ltd (China). The quality testing indexes of CHAC were analyzed as follows according to the Nut Shell Activated Carbon Test Standard (GB/T 7702-1997) of the National Standard of China: 0.4–3 mm particle size; 900–1,100 mg/g iodine value; 1,000 m<sup>2</sup>/g specific surface area; ≥90% strength; 0.45–0.55 cm<sup>3</sup>/g packing density; 100–150 mg/g methylene blue value; 8–10 pH; ≤5% moisture; and ≤9–12% ash. The CHAC powder was chosen as an adsorbent for subsequent sorption experiments without further treatment.

A stock solution of U(VI) (1,000 mg/L) was prepared by dissolving U<sub>3</sub>O<sub>8</sub> in a mixture of HCl, H<sub>2</sub>O<sub>2</sub>, and HNO<sub>3</sub>. U<sub>3</sub>O<sub>8</sub> was provided by the School of Nuclear Resources and Nuclear Fuel Engineering, University of South China. All working solutions of different U(VI) concentrations were obtained by diluting the stock solution with distilled water at room temperature. All chemical reagents used in this research were of analytical grade.

### 2.2. Batch sorption experiments

Batch sorption experiments were conducted in a series of 250 mL conical flasks. A 50 mL U(VI) solution was mixed with a known amount of CHAC powder. The pH of the solutions was adjusted to a desired value using 1.0 mol/L NaOH or HCl before mixing with the adsorbent. Thereafter, the flasks were agitated at 140 r/min on a rotary shaker. A sample of the solution was collected at suitable time intervals and filtered through a 0.45 µm membrane filter that does not adsorb uranyl cations. The filtrates were then analyzed for U(VI) concentration in the supernatants using a standard method given by Xie et al. [19]. The U(VI) removal efficiency and adsorption capacity of CHAC powder were calculated using the following equations:

$$Ad \% = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

$$Q_t = \frac{(C_0 - C_t) \times V}{W} \quad (2)$$

$$Q_e = \frac{(C_0 - C_e) \times V}{W} \quad (3)$$

where  $Ad$  % is the U(VI) removal efficiency;  $Q_e$  and  $Q_t$  are the adsorption capacity (mg/g) at equilibrium and at time  $t$ , respectively;  $C_0$ ,  $C_t$ , and  $C_e$  are the initial concentration, liquid-phase concentration at time  $t$ , and U(VI) equilibrium concentration (mg/L), respectively;  $V$  is the volume of the aqueous solution (L); and  $W$  is the mass of the adsorbent (g). All experiments were repeated twice and arithmetic average values were reported. Blank experiments were also conducted to ensure that no adsorption occurred on the walls of the glassware.

### 2.3. Equilibrium modeling

The Langmuir and Freundlich adsorption models were employed to correlate the obtained isotherm data. The relative parameters could be determined by linear regression analysis via Origin 8.0. The linearized Langmuir equation can be described as follows:

$$\frac{C_e}{Q_e} = \frac{1}{Q_{\max}} C_e + \frac{1}{bQ_{\max}} \quad (4)$$

where  $Q_e$  is the equilibrium adsorption capacity (mg/g);  $Q_{\max}$  is the maximum monolayer adsorption capacity (mg/g);  $C_e$  is the equilibrium concentration of U(VI) (mg/L); and  $b$  is the affinity constant related to the energy of adsorption (L/mg).

The linearized empirical Freundlich equation can be expressed as follows:

$$\ln Q_e = \ln K_F + \frac{1}{n} \ln C_e \quad (5)$$

where  $Q_e$  represents the equilibrium adsorption capacity (mg/g),  $C_e$  represents the equilibrium concentration of U(VI) (mg/L),  $K_F$  represents the Freundlich constant related to the adsorption capacity of the sorbent (mg/g), and  $n$  represents the Freundlich exponent related to adsorption intensity (dimensionless).

### 2.4. Kinetic modeling

To examine the kinetics of U(VI) uptake on CHAC powder, the pseudo-first-order and pseudo-second-order

kinetic models were used to analyze the adsorption kinetics. The nonlinear form of the pseudo-first-order equation is generally expressed as follows:

$$Q_t = Q_e(1 - e^{-k_1 t}) \quad (6)$$

where  $Q_e$  and  $Q_t$  are the adsorption capacity (mg/g) at equilibrium and at time  $t$  (min), respectively;  $k_1$  is the rate constant of the pseudo-first-order sorption ( $\text{min}^{-1}$ ). The values of the rate constant  $k_1$  and  $Q_e$  for the pseudo-first-order sorption reaction could be obtained by plotting  $Q_t$  versus  $t$  and by conducting further nonlinear regression analysis.

The linear form of the pseudo-second-order equation of Lagergren can be expressed as follows:

$$\frac{t}{Q_t} = \frac{t}{Q_e} + \frac{1}{k_2 Q_e^2} \quad (7)$$

where  $Q_e$  and  $Q_t$  are the adsorption capacity (mg/g) at equilibrium and at time  $t$ , respectively;  $k_2$  is the rate constant of pseudo-second-order sorption [g/(mg min)]. The values of rate constant  $k_2$  and  $Q_e$  for the pseudo-second-order sorption reaction could be determined by plotting  $t/Q_t$  versus  $t$  and by conducting further linear regression analysis.

## 3. Results and discussion

### 3.1. Effect of contact time

Contact time is a key factor that reflects adsorption kinetics. The effect of contact time on the adsorption of U(VI) on CHAC is shown in Fig. 1. The adsorption

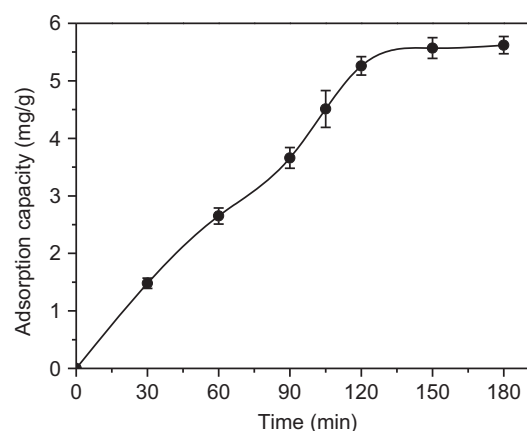


Fig. 1. Time course of U(VI) adsorption by CHAC (temperature: 25°C; pH: 2.0; U(VI): 200 mg/L; CHAC mass: 2.0 g; and solution volume: 50 mL).

capacity increased significantly within the initial 120 min, and a saturation level was gradually achieved at approximately 150 min. The amount of adsorbed uranyl cations changed insignificantly with a further increase in contact time. This phenomenon was due to the many vacant surface sites that were available for adsorption at an early stage. Near the equilibrium state, the residual vacant surface sites could not be occupied easily because of the repulsive forces between the solute molecules on the solid phase and bulk liquid phase. Thus, a contact time of 150 min was chosen in subsequent sorption experiments to guarantee an optimum U(VI) uptake.

### 3.2. Effect of pH

The effect of pH on the adsorption process is significant because pH affects the solution chemistry of metals, the activity of functional groups on the material surface, and the competition of metal ions. Uranium exists in different forms depending on the pH level. The U(VI) species distribution was determined under a wide variety of studied pH values using MINTEQ 3.0. At  $\text{pH} \leq 3.5$ , uranium existed predominantly as monomeric species ( $\text{UO}_2^{2+}$ ) with more than 95% content (Fig. 2). The hydrolyzed ionic species of uranium gradually increased with increasing pH. At  $\text{pH} \geq 5.0$ , uranium occurred mainly in the form of mononuclear and multinuclear ions, such as  $[(\text{UO}_2)(\text{OH})]^+$ ,  $[(\text{UO}_2)_2(\text{OH})_2]^{2+}$ ,  $[(\text{UO}_2)_3(\text{OH})_5]^+$ , and  $[(\text{UO}_2)_4(\text{OH})_7]^+$ .

The effect of initial pH on the percentage of U(VI) removal was examined at different pH levels ranging from 2.0 to 7.0 (Fig. 3). An acid condition was not helpful to U(VI) adsorption and the U(VI) removed was  $<40\%$  between pH 2.0 and 4.0. When the pH

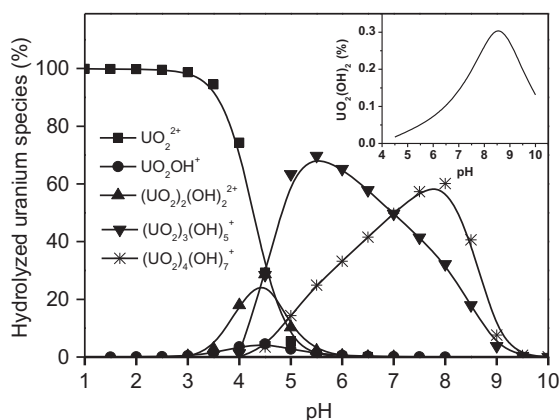


Fig. 2. Chemical speciation of hydrolyzed uranyl ions as a function of pH in pure water at 25°C and uranium concentration of 200 mg/L.

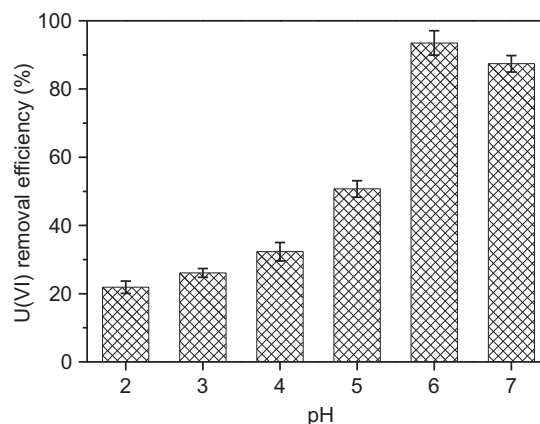


Fig. 3. Effect of pH on U(VI) adsorption by CHAC (temperature: 25°C; contact time: 150 min; U(VI): 200 mg/L; CHAC mass: 2.0 g; and solution volume: 50 mL).

moved toward neutral, the percentage of U(VI) removal increased markedly. CHAC showed a relatively good ability to adsorb U(VI) at near-neutral pH. The percentage of U(VI) removal reached a maximum value of 93.5% at pH 6.0. These results may be interpreted as follows.

When the pH was low, divalent free uranyl cations ( $\text{UO}_2^{2+}$ ) predominate in the solution. When the pH further increased, the percentage of ( $\text{UO}_2^{2+}$ ) in the solution declined, whereas the proportion of monovalent hydrolyzed species,  $\text{UO}_2(\text{OH})^+$ ,  $(\text{UO}_2)_3(\text{OH})_5^+$ , and  $[(\text{UO}_2)_4(\text{OH})_7]^+$  increased. Because these monovalent cations display much higher affinity to CHAC surface in ion exchange with protons, they may take the place of single protons on separate binding sites on the CHAC surface. Furthermore, since the ( $\text{UO}_2^{2+}$ ) is in a divalent state, it can solely replace two protons on the neighboring binding sites of the sorbent but cannot interact with those sites which are farther apart from one another. Hence, increasing the pH from 2.0 to 6.0 was favorable to U(VI) adsorption owing to an increment of various monovalent ions. As the pH exceeded 6.0 and moved toward 7.0, two kinds of precipitates, uranyl hydroxide ( $\text{UO}_2(\text{OH})_2$ ) and schoepite ( $\text{UO}_3 \cdot 2\text{H}_2\text{O}$ ), formed reducing the dissolved U(VI) levels in solution [20]. In this case, the U(VI) uptake might be partly hindered or interfered by the decrease of the U(VI) concentration available for adsorption. Therefore, the U(VI) removal efficiency slightly decreased at pH 7.0.

### 3.3. Effect of CHAC dosage

Adsorbent dosage is an important factor affecting the adsorption process since it determines the

adsorption capacity of an adsorbent for a given initial concentration of the adsorbate. The removal efficiency and adsorption capacity of U(VI) onto CHAC at six different sorbent dosages ranging from 0.25 to 2.5 g are shown in Fig. 4. Obviously, U(VI) removal efficiency increased with increasing adsorbent dosage up to 2.5 g at a fixed initial U(VI) concentration of 200 mg/L. Besides, an increase of adsorbent from 2.0 to 2.5 g led to an insignificant change in the U(VI) removal efficiency. By contrast, the adsorption capacity of CHAC decreased with increasing CHAC dosage. The adsorbent concentration effect was expected because an increasing adsorbent dose for a fixed initial metal concentration provided a high surface area or number of available adsorption sites for metal adsorption [21]. Within a certain range of initial U(VI) concentration, the percentage removal of U(VI) depended on the adsorption capacity of CHAC. The maximum U(VI) removal (96.5%) was obtained with an adsorbent dose of 2.5 g. In this study, 2.0 g of CHAC was chosen as the optimum adsorbent dosage for all further experiments because of this dosage provided a relatively high adsorption efficiency and acceptable adsorption capacity.

### 3.4. Effect of initial uranium concentration

The effect of initial U(VI) concentration on removal efficiency is presented in Fig. 5. U(VI) removal efficiency decreased with increasing initial U(VI) concentration. When the initial U(VI) concentration increased from 40 to 300 mg/L, the U(VI) removal efficiency decreased from 87.2% to 26.2%. In the U(VI) concentration range of 40–200 mg/L, the uranium adsorption

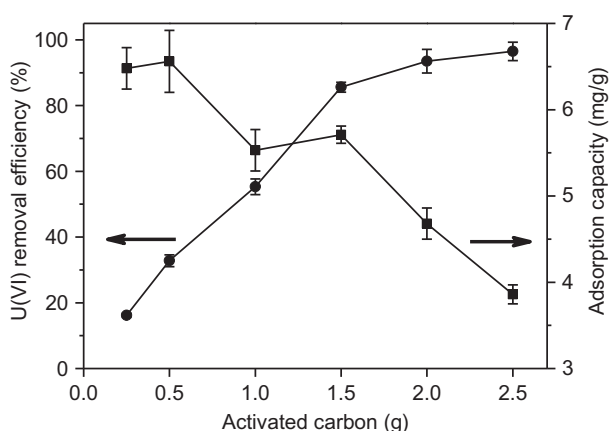


Fig. 4. Effect of CHAC dosage on U(VI) adsorption by CHAC (temperature: 25°C; contact time: 150 min; pH: 6.0; U(VI): 200 mg/L; and solution volume: 50 mL).

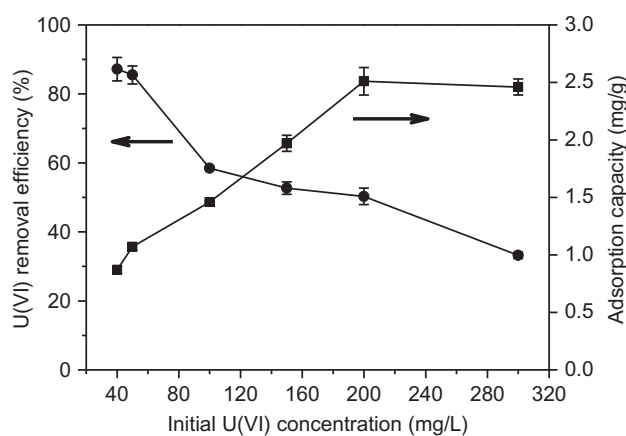


Fig. 5. Effect of initial U(VI) concentration on U(VI) adsorption by CHAC (temperature: 25°C; contact time: 150 min; pH: 5.0; CHAC mass: 2.0 g; and solution volume: 50 mL).

capacity increased with the increasing initial U(VI) concentration. This increase might be due to the high likelihood of collision between the metal ions and sorbent particles, which is driven by concentration gradients with increasing initial U(VI) concentration. However, when the initial U(VI) concentration was >200 mg/L, the uranium adsorption capacity remained nearly constant even though U(VI) concentration further increased. This finding indicated that sorption sites were saturated because only a limited number of surface sorption sites were available at high U(VI) concentrations.

### 3.5. Sorption isotherms

Adsorption isotherms describe the adsorbate–adsorbent interactions and play a crucial role in optimizing the use of adsorbents. Thus, the correlation of equilibrium data using empirical equations is important to the practical design and operation of adsorption systems. The Freundlich and Langmuir equations are the two most common mathematical models used to describe the adsorption isotherm. The Langmuir model is based on the assumptions of adsorption homogeneity, such as equally available adsorption sites, monolayer surface coverage, and nonexistent interaction between adsorbate molecules on adjacent sites [22]. The Freundlich isotherm can be employed for nonideal sorption on heterogeneous surfaces and multilayer sorption [23].

The calculated results of the Langmuir and Freundlich isotherm constants are listed in Table 1. The correlation coefficients of the Langmuir isotherm ( $R_L^2 = 0.991, 0.983$ ) were not only near 0.99 but greater

Table 1  
Adsorption equilibrium constants obtained from the Langmuir and Freundlich isotherms

Source of fitted data for deriving adsorption isotherm constants	Langmuir			Freundlich		
	$Q_{\max}$ (mg/g)	$b$ (L/mg)	$R^2$	$K_F$ (mg g <sup>-1</sup> (L mg) <sup>1/n</sup> )	$n$	$R^2$
Fig. 4	6.67	0.147	0.991	3.123	6.849	0.836
Fig. 5	2.08	0.189	0.983	0.626	4.000	0.815

Table 2  
Pseudo-first-order and pseudo-second-order adsorption rate constants for uranium sorption by CHAC

Pseudo-first-order			Pseudo-second-order			Experimental value $Q_{\text{exp}}$ (mg/g)
$k_1$ (min <sup>-1</sup> )	$Q_{e1}$ (mg/g)	$R_1^2$	$k_2$ (g/(mg min))	$Q_{e2}$ (mg/g)	$R_2^2$	
$7 \times 10^{-3}$	8.29	0.988	$2.43 \times 10^{-4}$	15.15	0.762	5.6

than those of the Freundlich isotherm ( $R_F^2 = 0.836$ , 0.815). The value of  $R^2$  indicated that the Langmuir isotherm model provided a better fit with the experimental data than the Freundlich isotherm model. Thus, the entire adsorption process followed a monolayer coverage.

The maximum uptake capacity  $Q_{\max}$  is an important parameter in evaluating the ability of adsorbents to accumulate U(VI) from aqueous solutions. The  $Q_{\max}$  value for uranium uptake by CHAC in this study was 6.67 mg/g. El-Sayed [24] reported that traditional wood activated carbon has a maximum U(VI) adsorption capacity of 30.95  $\mu\text{g/g}$ . In our previous research, we also reported that the  $Q_{\max}$  value for U(VI) uptake by apricot shell activated carbon is 59.17 mg/g [25]. Therefore, CHAC is a better adsorbent for uranium removal compared with conventional wood activated carbon, but its adsorption performance is inferior to that of apricot shell activated carbon.

### 3.6. Kinetic modeling

To examine the controlling mechanism of adsorption processes, such as mass transfer and chemical reaction, pseudo-first-order and pseudo-second-order kinetic equations were used to simulate U(VI) adsorption onto CHAC. The parameters of two fittings, including the kinetic constant ( $k$ ), correlation coefficient ( $R^2$ ), and equilibrium adsorption capacity ( $Q_e$ ), are listed in Table 2.

The correlation coefficient for the pseudo-second-order kinetic model ( $R_2^2 = 0.762$ ) was low. Furthermore, a large difference in the equilibrium adsorption

capacity ( $Q_e$ ) between the experimental value (5.6 mg/L) and calculation value (15.15 mg/L) was observed, thus indicating a poor pseudo-second-order fit to the experimental data. On the contrary, the plot of  $Q_t$  vs.  $t$  for the pseudo-first-order model gave a better correlation ( $R_1^2 = 0.988$ ) compared with the pseudo-second-order plot. The theoretical value of  $Q_e$  (5.6 mg/L) also agreed relatively well with the experimental value (8.29 mg/L). These facts suggest that adsorption of uranyl ions by CHAC follows the pseudo-first-order kinetic model rather than the pseudo-second-order kinetic model. A similar result was reported by Barkleit et al. [21] while working on U(VI) using conventional wood activated carbon.

## 4. Conclusions

The goal of this paper was to study the ability of CHAC to adsorb U(VI) ions from aqueous solutions. Under batch conditions, the U(VI) uptake process reached an equilibrium state after 150 min. The adsorption performance of CHAC was influenced by solution pH, sorbent dose, and initial U(VI) concentration. The optimum pH favorable for U(VI) adsorption is pH 6.0. The percentage of U(VI) removal increased with increasing CHAC dosage but decreased with decreasing initial adsorbate concentration. Higher initial U(VI) concentration resulted in higher U(VI) adsorption capacity for a given amount of CHAC. The isotherm data could be described well by the Langmuir model with the maximum monolayer adsorption capacity of 6.67 mg/g. The adsorption kinetics could be well-defined by the pseudo-first-order model. The

results of this research showed that CHAC could be considered an effective and available adsorbent for removing U(VI) from aqueous solutions.

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