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A Study of Removal Efficiency and Kinetics of Cadmium Using Activated Carbon Derived from Sulfurized Wastewater Treatment Plant Sludge

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A B S T R A C T

Cadmium (Cd) pollution in water bodies requires effective treatment methods that utilize sustainable alternative materials. This study aims to analyse the efficiency of Cd removal using sulfur-modified activated carbon derived from wastewater treatment plant (WWTP) sludge at varying contact times and to analyse its adsorption kinetics. We converted wastewater treatment plant (WWTP) sludge into activated carbon via KOH activation and Na₂S sulfurization. We conducted batch adsorption experiments at different contact times to evaluate Cd removal. Cd(II) removal reached 92.37% within 90 min (initial concentration: 20.32 mg/L, dosage: 2.5 g/L). The removal efficiency increased with contact time and remained relatively stable after 90 min, indicating that adsorption equilibrium had been achieved. Adsorption kinetics followed the pseudo-second-order model ($R^2 = 0.9845$), indicating chemisorption driven by sulfur surface sites. Sulfurization modified the chemical properties of the activated carbon surface and enhanced its affinity for Cd. These findings demonstrate the potential of sulfur-modified activated carbon derived from WWTP sludge as a sustainable adsorbent for Cd-contaminated water.

Contribution to Sustainable Development Goals (SDGs):

SDG 6: Clean Water and Sanitation

SDG 9: Industry, Innovation, and Infrastructure

SDG 12: Responsible Consumption and Production

1. INTRODUCTION

1.1. Research Background

Heavy metal contamination in aquatic environments is an environmental concern because these contaminants are toxic, persistent, and non-biodegradable. Cadmium (Cd) is one of the heavy metals of particular concern because its accumulation in aquatic ecosystems can adversely affect organisms and human health. Therefore, effective treatment technologies are required to reduce Cd concentrations in contaminated water. Adsorption is widely investigated as a treatment method because of its relatively simple operation and potential for using low-cost adsorbent materials [1].

The utilization of sewage sludge from wastewater treatment plants (WWTPs) as a precursor for activated carbon offers a *waste-to-resource* approach for pollutant removal. Sewage sludge contains carbonaceous components that can be converted into carbon-based adsorbents through carbonization and activation. The utilization of sewage sludge as an adsorbent precursor can simultaneously address sludge management issues and provide an alternative material for wastewater treatment, supporting the principles of a circular economy[1].

Surface modification through sulfurization can further enhance the interaction between activated carbon and metal ions. Previous studies have reported that sulfur modification can improve Cd removal compared with unmodified activated carbon by introducing sulfur-containing sites onto the adsorbent surface. [2]. In addition to surface modification, contact time is an important parameter in adsorption because it determines the



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opportunity for Cd^{2+} ions to diffuse toward the adsorbent surface and interact with available active sites. Kinetic analysis is therefore required to describe the adsorption rate and determine the kinetic model that best represents the Cd adsorption process [1].

Based on these considerations, this study aimed to analyse the Cd removal efficiency of sulfur-modified sewage sludge-based activated carbon at different contact times and to analyse the adsorption kinetics using the pseudo-first-order and pseudo-second-order models.

1.2. Literature Review

1.2.1. Adsorption

Adsorption is the process by which molecules, ions, or atoms attach to a surface. Adsorption is the process of capturing a substance from a fluid, whether a liquid or a gas, in the interfacial region, where the substance to be separated becomes bound to the surface of a solid material. This process forms a thin layer of adsorbate (the absorbed component) from the liquid phase (solvent) on the surface of the adsorbent (the absorbing substance) in the solid phase [3].

1.2.2. Activated Carbon

Activated carbon is a porous carbon material with a high surface area and many active sites, making it widely used as an adsorbent to remove various contaminants from water. Activated carbon can be produced from various carbon-rich materials, including wastewater treatment plant sludge. Using wastewater treatment plant sludge as a raw material for activated carbon can provide an alternative for waste management while producing a valuable material for water treatment. Carbonisation and activation processes, particularly chemical activation using KOH, can develop the pore structure and enhance the carbon's surface characteristics, thereby supporting heavy-metal adsorption. Previous research has shown that activated carbon derived from WWTP sludge can effectively remove Cd from solutions [2].

1.2.3. Sulfurization

Sulfurization is a surface modification method that introduces sulfur-containing species or groups onto the surface of activated carbon. This modification aims to enhance the adsorbent's surface affinity for metal ions by forming active sites better suited to interact with metals. For activated carbon derived from wastewater treatment plant sludge, the sulfurization process has been reported to improve Cd removal efficiency even though the adsorbent's specific surface area decreases, indicating that enhanced adsorption capacity is determined not only by physical characteristics but also by surface chemical properties [2].

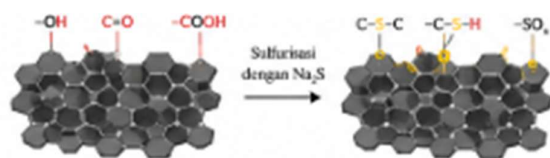


Figure 1 Changes in the Surface of Activated Carbon Before and After Sulfurization

Activated carbon derived from wastewater treatment plant sludge, which has undergone carbonization and KOH activation, has a porous carbon structure with various active sites on its

surface. These sites can take the form of oxygen-containing groups, such as hydroxyl ($-\text{OH}$), carbonyl ($\text{C}=\text{O}$), and carboxyl ($-\text{COOH}$), which interact with metal ions. The pore structure formed during activation also provides a surface area that allows Cd ions to diffuse and interact with the adsorbent surface. However, the surface chemistry of activated carbon before modification has not been specifically enriched with sulfur-donor sites that bind metal ions [3].

Following sulfurization with Na_2S , the activated carbon surface undergoes chemical modification by introducing sulfur-containing species. Surface sulfur can provide electron-donor sites that interact more strongly with Cd ions. Conceptually, sulfurization can enhance the affinity of the carbon surface for Cd through chemical interactions between Cd centers and sulfur-containing sites. Thus, sulfidation not only modifies the surface's chemical properties but also enhances the activated carbon's ability to capture Cd from solution [3].

1.2.4. Removal Efficiency

Removal efficiency is a parameter used to indicate an adsorbent's ability to reduce the concentration of contaminants in a solution following the adsorption process. For Cd removal, removal efficiency is determined by comparing the Cd concentration before and after adsorption. A higher efficiency value indicates a greater proportion of Cd successfully transferred from the solution phase to the adsorbent surface. Removal efficiency is influenced by various operating conditions, including contact time, because longer contact between the adsorbent and the adsorbate provides more opportunity for Cd ions to diffuse and interact with active sites. Research on activated carbon derived from wastewater treatment plant sludge shows that operating conditions, including contact time, affect the removal rate of heavy metals [3].

1.2.5. Adsorption kinetics

Adsorption kinetics describe the rate of the adsorption process and changes in the amount of adsorbate bound to the adsorbent surface over contact time. In this study, kinetic analysis was used to determine the most appropriate model for describing the removal of Cd ions by sulfur-modified activated carbon derived from wastewater treatment plant sludge. The adsorption capacity at each contact time (q_t) was calculated using the following equation:

$$q_t = \frac{C_0 \left(\frac{\text{mg}}{\text{L}} \right) - C_t \left(\frac{\text{mg}}{\text{L}} \right)}{m} \times V \quad (1)$$

Where q_t is the adsorption capacity at time t (mg/g), C_0 is the initial concentration of Cd (mg/L), C_t is the concentration of Cd at time t (mg/L), V is the volume of the solution (L), and m is the mass of the adsorbent (g) [3].

Mathematically, the first-order Lagergren model can be expressed as follows :

$$\text{Log}(q_e - q_t) = \text{Log } q_e - \frac{k_1}{2,303} t \quad (2)$$

Where :

q_e = Amount of ions absorbed at equilibrium (mg/g)

q_t = Amount of ions absorbed at time t (mg/g)

k_1 = Adsorption rate constant (min^{-1})

From the above equation, the adsorption rate constant (K_1) can be determined by performing a linear regression of the $\text{log}(q_e - q_t)$ function against time (t), whereas the pseudo-second-order

equation is based on the solid-phase adsorption rate, which is expressed in the following equation [3].

$$\frac{t}{qt} = \frac{1}{k_2 qe^2} + \frac{1}{qe} t \quad (3)$$

1.3. Research Objective

This study aims to analyze the Cd removal efficiency of sulfur-modified activated carbon derived from sewage sludge at different contact times and to analyze the adsorption kinetics using the pseudo-first-order and pseudo-second-order models.

2. MATERIALS AND METHODS

Preliminary testing showed that the WWTP sludge used as the adsorbent raw material contained no detectable cadmium (Cd), with a measured concentration of 0.00 mg/g. The analysis was conducted using the Atomic Absorption Spectrophotometry (AAS) method at a KAN-accredited laboratory, confirming that the sludge was free from Cd contamination prior to the carbonization, activation, and sulfidation processes.

2.1. Preparation of Activated Carbon from Sewage Treatment Plant Sludge

The preparation process for activated carbon derived from wastewater treatment plant sludge followed previous research. Raw wastewater treatment plant sludge was collected from a belt filter press at an industrial tobacco wastewater treatment plant in Pasuruan, then washed with deionized water and dried in an oven at 105°C. The dried sludge was ground to a particle size of 0.1 mm, then carbonized at 712.7°C for 2 hours. The carbonized material was subsequently activated using KOH at a mass ratio of wastewater treatment plant sludge to KOH of 1:1 and impregnated for 24 hours. After the impregnation process, the carbon was washed with deionized water until the pH reached 7 and dried at 105°C. The resulting activated carbon was then modified by sulfidation using a 1.56 M Na₂S solution for 24 hours. After impregnation, the adsorbent was washed with deionized water until the pH reached 7 and dried at 105°C [2].

2.2. Batch Adsorption Study and Data Analysis

Cd adsorption was performed using the batch adsorption method by adding 2.5 g/L of sulfurized activated carbon derived from wastewater treatment plant sludge to 100 mL of a Cd solution with an initial concentration (C₀) of 20.32 mg/L. The mixture was shaken at 150 rpm for 5, 10, 20, 30, 40, 60, 80, 90, 100, and 120 minutes. After the adsorption process, the mixture was separated to obtain the filtrate, and the final Cd concentration (C_t) was analyzed using an Atomic Absorption Spectrophotometer (AAS). The Cd concentration data before and after adsorption were used to determine the Cd removal efficiency using the following equation:

$$\text{Removal Efficiency (\%)} = \frac{C_0 \left(\frac{\text{mg}}{\text{L}} \right) - C_t \left(\frac{\text{mg}}{\text{L}} \right)}{C_0 \left(\frac{\text{mg}}{\text{L}} \right)} \times 100 \quad (4)$$

In addition, contact time data were used to calculate the adsorption capacity (qt) and to analyze the adsorption kinetics using the pseudo-first-order and pseudo-second-order models [3].

3. DISCUSSION

3.1. Cadmium Removal Efficiency

Cadmium (Cd) removal efficiency is the primary parameter used to evaluate the ability of activated carbon derived from wastewater treatment plant sludge to reduce Cd concentrations in solution. Removal efficiency is determined by the difference in Cd concentration before and after adsorption, namely the initial concentration (C₀) and the final concentration (C_e). The greater the reduction in Cd concentration after the adsorption process, the higher the removal efficiency, which indicates the adsorbent's ability to bind Cd from the solution [3]. The following table presents Cd removal efficiency results at various contact times.

Table 1. Cadmium Removal Efficiency

Contact Time (Minutes)	Initial Concentration (mg/L)	Final Concentration (mg/L)	Removal Efficiency (%)
5	20.32	3.02	85.13
10		2.79	86.26
20		2.56	87.40
30		2.40	88.18
40		2.17	89.32
60		2.02	90.05
80		1.78	91.24
90		1.55	92.37
100		1.58	92.22
120		1.59	92.17

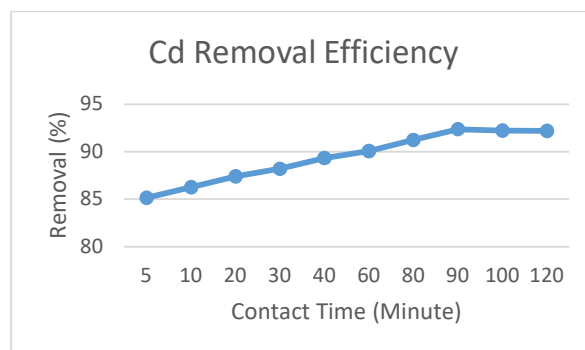


Fig. 2 Cadmium Removal Efficiency

Based on the table and Figure 2 above, the Cd removal efficiency increased with increasing contact time, from 85.13% at 5 min to 92.37% at 90 min. The rapid increase in removal efficiency during the initial stage is attributed to the abundant active sites on the activated carbon surface and the large concentration gradient between Cd ions in solution and the adsorbent surface, which promotes rapid Cd binding to the surface [3].

As contact time increased, removal efficiency increased more gradually because available surface active sites became increasingly occupied by Cd ions. At this stage, Cd ions progressively diffuse into the pores of the activated carbon, resulting in a slower adsorption rate. At 90 min, the Cd removal efficiency reached the highest value of 92.37% and remained relatively stable thereafter, indicating that the adsorption system had approached equilibrium [4]. Thus, the adsorption process can be described as rapid initial surface binding followed by slower

pore diffusion as available active sites become increasingly occupied.

3.2. Adsorption Kinetics

Kinetic analysis was conducted to determine the behavior and rate of Cd adsorption by activated carbon derived from sulfurized wastewater treatment plant sludge at various contact times. The adsorption data were analyzed using the pseudo-first-order (PFO) and pseudo-second-order (PSO) models. Both models were used to determine the most appropriate kinetic model for describing the relationship between contact time and the Cd adsorption process. The kinetic parameters obtained included the theoretical adsorption capacity (q_e), the adsorption rate constant (k), and the coefficient of determination (R^2) [3]. Table 2 presents the results of the analysis of the two kinetic models.

Table 2. Adsorption Kinetics Models

Model	Parameter	Results	Unit
PFO	q_e	1.643	mg/g
	K	0.153	min^{-1}
	R^2	0.508	
PSO	q_e	1.015	mg/g
	K	1.888	$\text{g/mg} \cdot \text{min}$
	R^2	0.9845	

Based on Table 2, the PFO model yielded a theoretical q_e value of 1.643 mg/g, a rate constant (k_1) of 0.153 min^{-1} , and an R^2 value of 0.508. Meanwhile, the PSO model yields a q_e value of 1.015 mg/g, a rate constant (k_2) of $1.888 \text{ g/mg} \cdot \text{min}$, and an R^2 value of 0.9845. The difference in R^2 values indicates that the PSO model fits the experimental data better than the PFO model. To illustrate the relationship between contact time and each kinetic model, the following Figure 3 presents the PFO and PSO linear regression results.

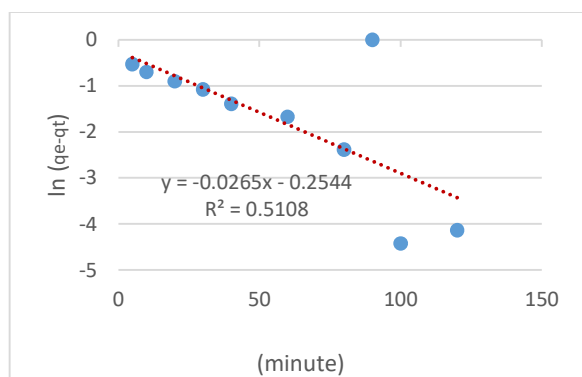


Fig. 3 Linear plot of Cd adsorption kinetics using the pseudo-first-order model

Based on the pseudo-first-order (PFO) graph, a linear regression equation of $y = -0.3525x - 0.2158$ was obtained, with an R^2 value of 0.508. This value indicates that the PFO model has a relatively low correlation with the Cd^{2+} adsorption data across varying contact times. This suggests that changes in adsorption capacity over time do not fully follow the assumptions of the PFO model. Thus, the PFO model is less capable of representing the kinetic characteristics of Cd adsorption on activated carbon derived from sulfidized wastewater treatment plant sludge.

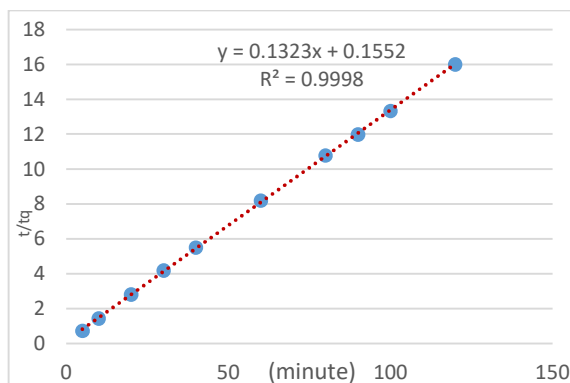


Fig. 4 Linear plot of Cd adsorption kinetics using the pseudo-second-order model

Based on Figure 4, the Pseudo-Second-Order (PSO) model produced a coefficient of determination (R^2) of 0.9845. This value was higher than that obtained for the Pseudo-First-Order (PFO) model, which was 0.508. The higher R^2 value indicates that the PSO model better describes changes in Cd adsorption capacity with contact time. Overall, the PSO model provides a better fit than the PFO model based on the R^2 value (0.9845 vs 0.508). These results indicate that the Cd adsorption process by activated carbon derived from sulfurized WWTP sludge is more accurately described using the pseudo-second-order model. The good fit of the PSO model suggests that interactions between Cd ions and active sites on the adsorbent surface contribute to adsorption. This can be attributed to sulfidation, which modifies the chemical properties of the activated carbon surface by adding sulfur-containing species. Sulfur-containing sites can increase surface affinity for Cd, facilitating interactions between metal ions and the adsorbent surface [3].

3.3. Cadmium Removal Mechanisms

The process of Cd removal by activated carbon derived from sulfurized wastewater treatment plant sludge results from changes in the adsorbent's surface characteristics that occur during the preparation stages, namely carbonization, activation, and sulfuration. Each stage plays a distinct role in the adsorbent's ability to interact with Cd. Carbonization converts the organic components of the sludge into a carbon structure, while activation using KOH helps develop the pore structure and provides surface area and active sites for the adsorption process [2]. Subsequently, sulfidation modifies the surface chemistry by introducing sulfur-containing species. Previous research on activated carbon derived from WWTP sludge has shown that sulfidation can introduce sulfur-containing groups onto the surface and improve the efficiency of heavy metal removal, including Cd.

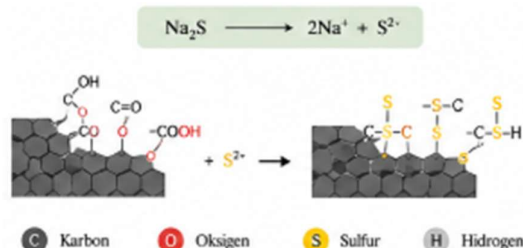


Fig. 5 Mechanism of Sulfur-Containing Active Site Formation

The sulfidation process using Na_2S can be described as sulfide species in solution interacting with the activated carbon surface and producing sulfur-containing surface sites. These sites can then donate electrons to Cd ions. Therefore, sulfidation can increase the adsorbent's surface affinity for Cd through stronger chemical interactions. Research on sulfured carbon materials also indicates that interactions between Cd and surface sulfur species can contribute to the Cd removal mechanism [3].

This relationship can be explained using the Hard and Soft Acids and Bases (HSAB) theory. The Cd ion exhibits soft acid characteristics, while the sulfur atom at the sulfur-containing sites exhibits relatively soft donor characteristics. Based on the HSAB principle, soft species tend to form stronger interactions with partners that are also soft. Thus, the presence of sulfur sites on sulfurized activated carbon can increase the tendency for interactions between Cd and the adsorbent surface. This interpretation is also consistent with studies on the sulfurization of activated carbon, which show an increase in the adsorbent's ability to bind Cd following sulfur modification [4].

This mechanism is further related to the effect of contact time on removal efficiency. In the early stages of adsorption, the Cd concentration in solution is still high and most active surface sites are available, so mass transfer and Cd interactions occur relatively quickly. As contact time increases, Cd occupies more active sites, and the adsorption rate gradually decreases. This trend is evident from the increase in removal efficiency from 85.13% at 5 minutes to 92.37% at 90 minutes. After 90 minutes, the efficiency remained relatively constant, at 92.22% at 100 minutes and 92.17% at 120 minutes, indicating that the system had approached equilibrium.

These results are also consistent with the kinetic analysis in this study. The pseudo-second-order (PSO) model yielded an R^2 value of 0.9845, higher than that of the pseudo-first-order model (0.508). The good fit of the PSO model indicates that interactions between Cd and active surface sites contribute to adsorption. Thus, the results regarding efficiency, contact time, and kinetics provide a consistent trend indicating that Cd^{2+} removal is not only related to mass transfer toward the carbon surface but also to the interaction of Cd with the sulfur-modified active sites [5].

Overall, the Cd removal mechanism can be viewed as a stepwise process that begins with the transport of Cd from the solution to the activated carbon surface, followed by diffusion to the active sites, and then interaction of Cd with the available surface sites. Carbonization and activation play a role in the formation of the carbon structure and pores, while sulfurization modifies the surface's chemical characteristics, thereby increasing its affinity for Cd^{2+} . Together, these physical and chemical characteristics yield a maximum removal efficiency of 92.37% at 90 minutes and a better kinetic fit with the PSO model.

4. CONCLUSION

This study shows that activated carbon derived from wastewater treatment plant sludge and modified through sulfidation is capable of effectively removing Cd from solution. Contact time influenced the adsorption process, with removal efficiency increasing to 92.37% at 90 minutes and then remaining relatively stable at 100–120 minutes. Kinetic analysis results indicate that the pseudo-second-order (PSO) model better describes the adsorption process than the pseudo-first-order (PFO) model, with R^2 values of 0.9845 and 0.508, respectively. The PSO fit suggests

that interactions between Cd and surface active sites contribute to adsorption. The carbonization and activation processes play a role in the formation of the carbon structure and active sites, while sulfurization using Na_2S modifies the surface chemistry through the presence of sulfur-containing species that have the potential to increase the affinity for Cd. The subsequent removal mechanism can be attributed to Cd migration toward the adsorbent surface and its interaction with active sites, including sulfur-based interactions based on the HSAB approach. Overall, sulfurized activated carbon derived from wastewater treatment plant sludge shows potential as an alternative adsorbent for Cd removal.

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