

Adsorptive Removal of Chromium (VI) Ions from Aqueous Solution by Banana Pseudo Stem Adsorbent

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ABSTRACT

The presence of Cr ions in wastewater must be treated before being released into the environment due to its detrimental impact on both the environment and human health. In this study, the removal of Cr (VI) ions from an aqueous solution was investigated by adsorption using an adsorbent derived from agriculture wastes, banana pseudo stem. The adsorbent was prepared by oven-drying the banana stem waste at 105°C for 24 h. The surface structure of the adsorbent was characterized using scanning electron microscopy (SEM). Batch adsorption experiments were carried out to determine the removal efficiency of Cr (VI) ions based on four adsorption operation parameters: pH of the solution, adsorbent dosage, contact time and initial concentration of ion solution. At room temperature, the highest Cr (VI) ions removal of 88.2% was achieved using 0.5 g banana pseudo stem adsorbent, with an initial concentration of chromium solution of 500 ppm at pH 2 and after 90 min of contact time. For the equilibrium study, the experimental data were better fitted by the Langmuir isotherm model with a maximum adsorption capacity of 33.33 mg/g. Meanwhile, the kinetic isotherm was best fitted by the pseudo-second-order model. Therefore, the banana pseudo stem showed great potential as an efficient, low-cost and natural green adsorbent for Cr (VI) ions removal from an aqueous solution via adsorption.

1. INTRODUCTION

Heavy metal ions are being excessively released from industrial waste. One of the harmful metal ions that can easily be found in water sources is chromium. In the aquatic environment, this metal primarily exists in the forms of trivalent Cr (III) ions and hexavalent Cr (VI) ions. Compared to Cr (III) ion, Cr (VI) ion, requires more attention due to its poisonous, high solubility, mutagenicity, and carcinogenicity (Aharchaou et al., 2018). In humans, excessive dosage of chromium can lead to serious health issues such as ulcers, lung cancer, diarrhoea, lip and nasal irritation, asthma and kidney failure (Stambulski et al., 2018). Chromium is easily bio-accumulated at low concentrations in aquatic organisms. According to Speer et al. (2019), exposure to Cr (VI) ion in fish changed hatching times, causing DNA damage and reducing reproduction rates. Thus, removing Cr (VI) ions from water is essential for the health of all living creatures and the environment.

To date, many conventional and modern methods have been employed in the treatment of heavy metals removal from water and wastewater. Adsorption has been the most prominent due to its high removal efficiency, economical, cheaply accessible, simple operation and environmentally friendly (Sukmana et al., 2021). Recently, researchers are paying more attention to utilizing agricultural waste as the adsorbent feedstock for treating heavy metals problems in wastewater via the adsorption method. Adsorbents derived from walnut (Garg et al., 2023), soybean straw (Guo et al., 2021), corncob (Yang et al., 2018), sugarcane bagasse (Abilio et al., 2021), wheat straw (Song et al., 2021), and coconut husk (Hanafiah et al., 2020) have been applied in the study of removing Cr (VI) ions from water. Due to its renewable nature, using adsorbent made from agricultural waste can be both environmentally friendly and more cost-effective.

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Banana (*Musa* spp.) is Malaysia's second-largest cultivated fruit after durian. It is estimated about four times waste is generated for every cycle of banana production (Taib, 2019). Massive banana waste definitely creates a serious disposal issue, however, it is also a significant resource waste. The presence of abundance of lignocellulose polymers with different functional groups, (mainly hydroxyl, carboxyl and carboxylate group) in the banana biomass creates effective adsorption with heavy metal ions, dyes and other pollutants. Banana wastes like fruit peels, leaves and stems were converted into biochar and have been tested as an adsorbent for dyes and heavy metals removal (Baharim et al., 2023; Kokate et al., 2022; Liu et al., 2022).

Earlier, several studies were focusing on the Cr ions removal from aqueous solution using adsorbent and biochar or modified biochar derived from banana wastes. Selimin et al. (2022) reported in their study using banana blossom peels adsorbent with chemical treatment was able to achieve a maximum of 227.27 mg/g Cr (VI) adsorption capacity. Research by Payel et al. (2020) and Xu et al. (2018) using banana rachis biochar (carbonized at 650°C) and banana pseudostem biochar produced at 500°C for chromium removal achieved maximum adsorption capacities at 2,500 mg/g and 43.47 mg/g, correspondingly. Meanwhile, banana straw biochar loaded with magnesium chloride showed a high adsorption performance of Cr (VI) ions, with 125.0 mg/g capacity as reported by Li et al. (2020).

Even though the adsorption efficiency of the adsorbents significantly improved, the facts showed that biochar or modified biochar is non-economical since involving with high energy consumption for the carbonization procedure and utilization of hazardous chemical solution for pretreatment. Studies using natural banana waste adsorbent in removing Cr (VI) ions are still very limited. Therefore, this present study utilizes banana pseudo stem waste as biomass feedstock for producing low-cost, natural and unmodified adsorbent in removing Cr (VI) from an aqueous solution. The adsorbent was prepared by a simple and easy drying technique and without any modification or additional chemical pretreatment. The effectiveness of the banana pseudo stems adsorbent in removing Cr (VI) ions from an aqueous solution was investigated in batch mode of adsorption experiments. The adsorption operation parameters influencing Cr (VI) removal efficiency were examined, involving solution pH, adsorbent dosage, initial metal ion concentration and contact time. The experimental

results were modelled using different isotherms and kinetic adsorption models.

2. METHODOLOGY

2.1 Materials

All chemicals used in this study were analytical grade (AR) reagents. 2.8287 g of $K_2Cr_2O_7$ (System) was dissolved with 1,000 mL of distilled water to prepare a 1,000 ppm Cr (VI) standard stock solution. The stock solution was diluted with distilled water to the required concentration for further use in the adsorption experiment. HCl and NaOH were purchased from Sigma-Aldrich and HmbG Chemicals, respectively.

2.2 Preparation of banana pseudo stem adsorbent

The raw material, banana pseudo stems of *Musa paradisiaca* (BPS) was collected from a banana farm in Ijok, Selangor, Malaysia. The stems were washed with tap water multiple times and then cut into small pieces. After that, the stems were air-dried for five days and continued to dry at 105°C for 24 h using an oven (Venticell 55, USA). After being cooled, the produced BPS adsorbent was ground into powder, sieved at 1 mm and finally kept in a container prior to further use.

2.3 Characterization of banana pseudo stem adsorbent

The surface morphology of the BPS adsorbent was observed using scanning electron microscopy (SEM) (Zeiss supra 40, Germany) operated at 10 kV with 5.00 K magnification.

2.4 Adsorption experiments

At room temperature, the effectiveness of BPS adsorbent in removing Cr (VI) ions from aqueous solution was investigated through batch adsorption tests to determine the effects of adsorption factors, including pH, adsorbent dosage, starting concentration, and contact time. The maximum adsorption conditions were determined by changing the parameter studied while the other parameters were kept constant, as detailed in Table 1. The experiments were performed with 100 mL of Cr (VI) ions solution in a 250 mL conical flask. Prior experiment, the Cr (VI) ions solution was adjusted to a specific pH value with 0.1 M HCl or 0.1 M NaOH and was measured with a pH meter (Sartorius PB-10, China). The mixtures were agitated at 150 rpm in a temperature-controlled shaker (PROTHERM, USA) for the required time. After adsorption, the mixture was

filtered and the absorbance of the Cr (VI) ions was measured with a complexing agent, 1,5-diphenyl-carbazide, at 350 nm wavelength using UV-Vis Spectrophotometer (Hitachi, Japan). Experiments were repeated three times, for the accuracy of the data analysis. The percentage removal, % R, of the Cr (VI) ions and the adsorption capacity, q_e , of the BPS were determined using the following equations.

$$\text{Percentage removal, \% R} = \left(\frac{C_0 - C_e}{C_0} \right) \times 100\% \quad (1)$$

$$\text{Adsorption capacity, } q_e = \left(\frac{C_0 - C_e}{m} \right) \times V \quad (2)$$

Where; C_0 is the initial concentration (ppm); C_e is the concentration at equilibrium (ppm); V is the volume (L); and m is the mass of the adsorbent (g).

2.5 Statistical analysis

The percentage removal of Cr (VI) ions was statistically analyzed using the IBM SPSS Statistics 27 software utilizing the Variance analysis (ANOVA) and Tukey's test for mean comparison at 95 % reliability ($p < 0.05$). All data were presented as mean \pm standard deviation (SD).

Table 1. Conditions of batch adsorption experiments, performed at 25°C

Adsorption parameters	pH range	Adsorbent dosage range (g)	Initial concentration range (ppm)	Contact time range (min)
Effect of pH	2, 3, 4, 5, 6, 7, 8, 9, 10	0.5	100	60
Effect of adsorbent dosage	2	0.1, 0.2, 0.3, 0.4, 0.5, 0.6	100	60
Effect of initial concentration	2	0.5	100, 200, 300, 400, 500	60
Effect of contact time	2	0.5	500	10, 20, 30, 60, 90, 120

3. RESULTS AND DISCUSSION

3.1 Characteristic of banana pseudo stem adsorbent

The SEM micrographs of BPS adsorbent obtained before and after Cr (VI) ions adsorption are shown in Figure 1. Figure 1(a) demonstrates that before adsorption, the BPS particles have bundle-

aligned fibres with rough lignocellulosic surface structures and uneven shapes, which can help the adsorption. Following the adsorption of metal ions, the micrograph in Figure 1(b) shows the microparticle adhesion to the fibre surface. Riguetto et al. (2021) who used banana pseudo stem in the adsorption of textile dye, also reported this same behaviour.

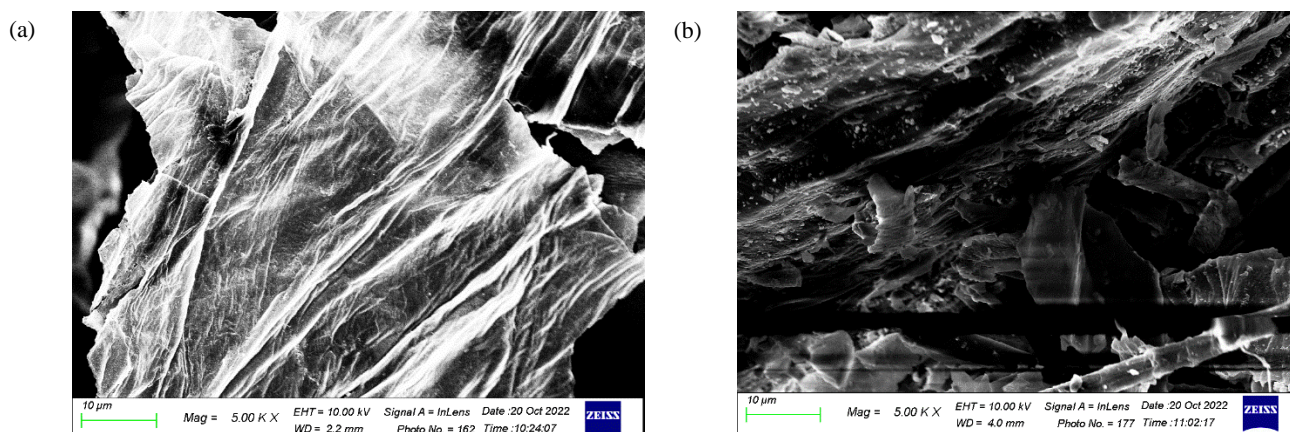


Figure 1. SEM micrograph of BPS adsorbent (a) before adsorption (b) after adsorption

3.2 Effect of adsorption experiments

The effect of four adsorption experiments is discussed in the following subsections.

3.2.1 Effect of pH

The pH of the solution affects the surface charges and the ionic state of functional groups on the

adsorbent surface (Birhanu et al., 2020). Figure 2 depicts the pH solution effect from pH 2 to 10 on the removal percentage of Cr (VI) ions using the BPS adsorbent. Generally, the removal efficiency declines as the solution pH increases. The highest removal of 49.3% was achieved at pH 2, and the removal

continuously decreased to 41.1% with the increase in pH to 10. Since high acidity condition creates stable cations and more formation of HCr_2O_7^- which was conducive to the electron interaction between the adsorbent and chromate anions, allowing for more removal via reduction (Qasem et al., 2021). In contrast, the solubility of metal cations declines with higher pH levels, raising the probability of a precipitation occurrence. Gupta et al. (2018) reported similar trends of the pH effect on Cr (VI) removal were reported by using treated corncob biochar, with pH 2 showing the highest 93.0% removal.

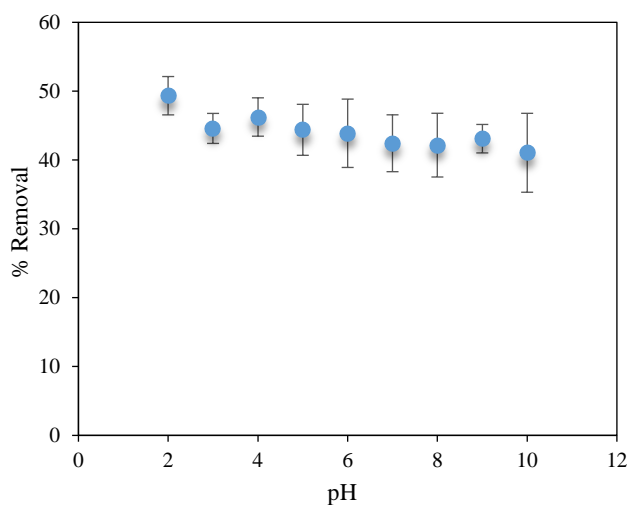


Figure 2. Effect of pH on the % removal of Cr (VI) ions

3.2.2 Effect of adsorbent dosage

The effect of adsorbent dosage on the Cr (VI) ions removal was examined by applying different dosages; 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6 g and the results are represented in Figure 3. The removal efficiency improved by 15.4% as the adsorbent increased from 0.1 to 0.4 g, and reached the highest removal of 46.1% with 0.5 g adsorbent dosage. There may be more surface area or active site and surface functional groups accessible for adsorption, which would explain the increase in Cr (VI) removal as adsorbent dosages increase (Garg et al., 2023). Further dosage caused no noticeable increase in the percentage removal. At this point, there were limited Cr (VI) ions in the aqueous solution which resulted in numerous unused active sites, thus the removal efficiency becoming constant or reducing. Similar adsorbent dosage effect findings on Cr (VI) ions removal have recently been published by Birhanu et al. (2020) and Bayuo et al. (2019) using groundnut shell and Ethiopian Oदारacha adsorbent, respectively.

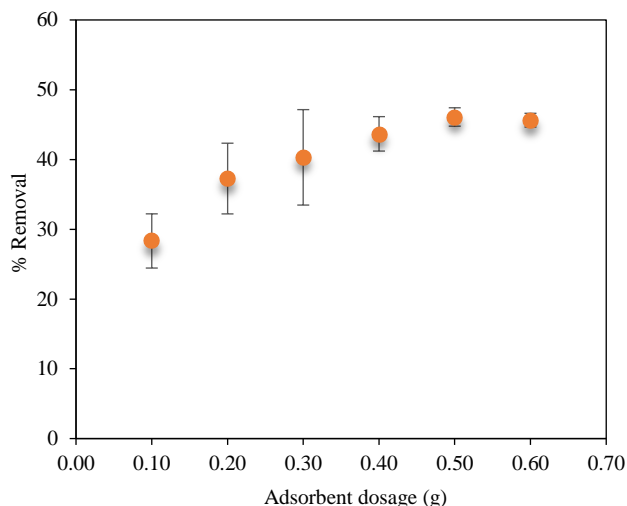


Figure 3. Effect of adsorbent dosage on the % removal of Cr (VI) ions

3.2.3 Effect of initial concentration

The initial Cr (VI) ions concentration plays an essential role as a driving force in reducing mass transfer resistance between the adsorbent's surface and the metal solution. Figure 4 shows the effect of different Cr (VI) initial concentrations on the removal efficiency. Cr (VI) ions removal was drastically increased by 35.0% with the increase in initial concentration from 100 ppm to 300 ppm. This observation might be caused by the adsorbent surface having a large number of active sites available, which would increase the Cr (VI) adsorption (Jock et al., 2021). The percentage removal continued to increase by 6.4% at 400 ppm and thereafter achieved the highest removal of 88.4% at 500 ppm. At 600 ppm, the removal efficiency of Cr (VI) decreases as insufficient availability of active sites, thus causing the saturation of the adsorbent's surface at equilibrium. Similar initial Cr (VI) concentration effects on removal trends were observed for adsorbents prepared from iron-based solid waste and bentonite clay by Qi et al. (2023) and Jock et al. (2021), respectively.

3.2.4 Effect of contact time

The effect of contact time was investigated in order to determine the equilibrium time for the percentage removal of Cr (VI) ions. The experiments were run with various contact times ranging from 10 min to 120 min, and the outcomes are shown in Figure 5. The trend of Cr (VI) removal was gradually increased by 1.5% from 10 min to 60 min. This might be explained by the adsorbent's numerous available active sites, which allow for efficient adsorption. Then, the percentage removal reached the highest of 91.9% at

90 min indicating the equilibrium of the adsorption was established. Further, a longer time period will hinder any additional adsorption due to the adsorbate's difficulty to find accessible unoccupied sites. Bayuo et al. (2019) reported a similar result on the contact time effect for the Cr (VI) removal using groundnut shell adsorbent. However, with lower maximum removal (81.6%) and a longer time for achieving equilibrium (120 min) as compared to this current study.

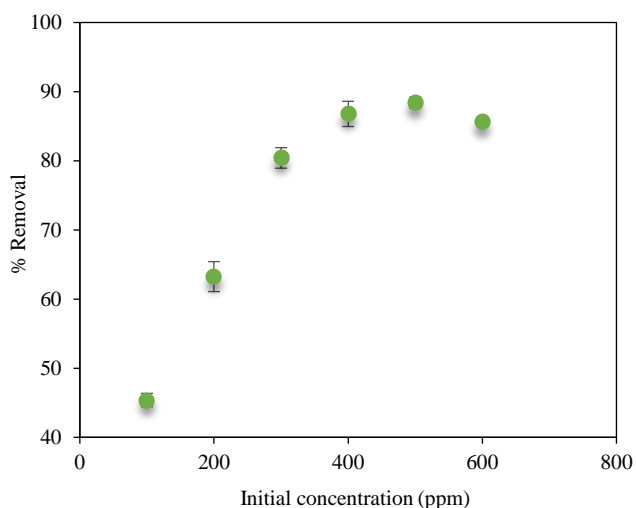


Figure 4. Effect of initial concentration on the % removal of Cr (VI) ions

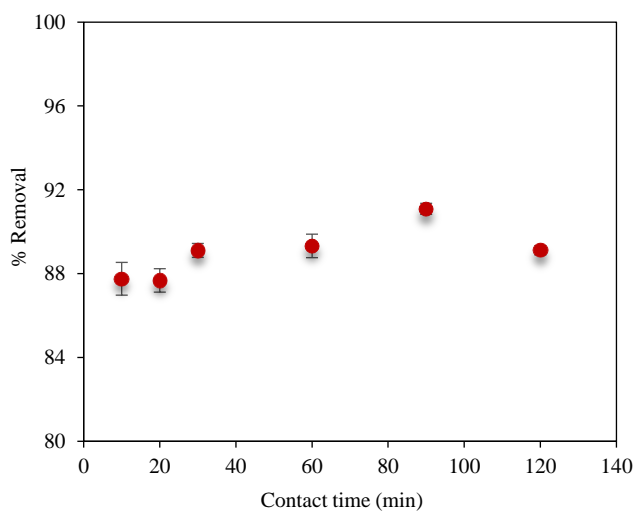


Figure 5. Effect of contact time on the % removal of Cr (VI) ions

3.3 Adsorption isotherm

Adsorption isotherm describes the adsorption distribution of Cr (VI) ions onto the surface of the BPS adsorbent solid phase of the biochar under equilibrium conditions. Two isotherm models; Langmuir and Freundlich were employed for the adsorption isotherm study. The Langmuir model assumes the adsorbate creates a monolayer on the homogenized adsorbent

surface and acts as an independent entity. Meanwhile, the Freundlich model is used to simulate multilayer adsorption on heterogeneous surfaces. The isotherms are expressed by the linear equation as follows:

$$\text{Langmuir: } \frac{1}{q_e} = \frac{1}{bC_e q_m} + \frac{1}{q_m} \quad (3)$$

$$\text{Freundlich: } \log q_e = \frac{1}{n} \log C_e + \log K_F \quad (4)$$

Where; q_e (mg/g) is the amount of Cr (VI) ions adsorbed per unit mass of adsorbent in equilibrium, C_e (mg/L) is the Cr (VI) ions concentration at equilibrium. q_m (mg/g) and b (L/mg) are the maximum monolayer adsorption capacity and Langmuir constant, respectively. Meanwhile, K_F ((mg/g) (L/mg)^{1/n}) is the Freundlich constant which is related to adsorption capacity and $1/n$ indicates the adsorption intensity. If $1/n=1$, the separation within the two phases is not dependent on the concentration. If the value of $1/n < 1$, it shows normal adsorption. If $1/n > 1$, it shows cooperative adsorption.

Figure 6 shows data from equilibrium adsorption tests fitted using both isotherm equations. By comparing the linear regression correlation coefficient, R^2 , the Langmuir model is better fitted to the experimental data, which concludes the monolayer adsorption of Cr (VI) ion onto the homogenous BPS adsorbent's surface. According to the Langmuir model, the maximal adsorption capacity is 33.33 mg/g. Bayuo et al. (2019) and Martín et al. (2016) reported similar adsorption isotherm results for the Cr (VI) ions removal using groundnut shells and Canary banana peels.

3.4 Adsorption kinetics

Adsorption kinetics provides crucial details regarding the reaction mechanism and the reaction's rate-limiting stage. This study applied two common models, pseudo-first-order and pseudo-second-order, to obtain the best-fitted kinetic model for the Cr (VI) ion's adsorption onto the adsorbent surface. Equations used to express the linearized pseudo-first-order and pseudo-second-order kinetic are given below:

$$\text{Pseudo-first-order: } \ln(q_e - q_t) = \ln q_e - k_1 t \quad (5)$$

$$\text{Pseudo-second-order: } \frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (6)$$

Where; q_e (mg/g) and q_t (mg/g) are the amount of Cr (VI) ions adsorbed in equilibrium and given time, respectively. k_1 (min⁻¹) and k_2 (g/mg·min) are the pseudo-first-order constant and pseudo-second-order constant, respectively.

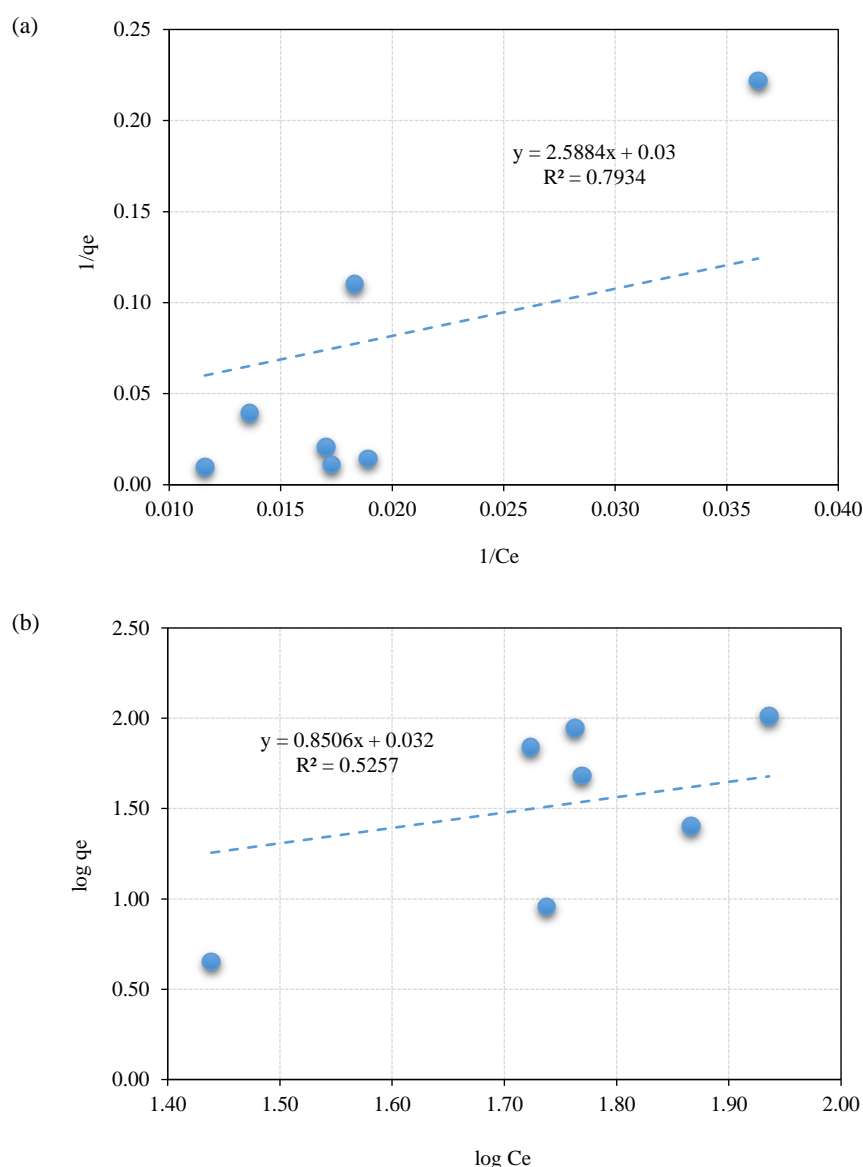


Figure 6. Linearized isotherm equation based on, (a) Langmuir; (b) Freundlich model for Cr (VI) ions adsorption

Figure 7 presents the experimental data results obtained in the kinetic study of Cr (VI) ions adsorption by BPS adsorbent. The correlation coefficient, R^2 , demonstrates that the pseudo-second-order model has better fitness than the pseudo-first-order model. This implies that adsorption is influenced by the way Cr (VI) ions interact with the adsorbent surface. Additionally, the outcomes demonstrate the presence of vacant adsorption sites on the banana stem surface significantly affects the adsorption rate as opposed to the number of metal ions adsorbed. This study's findings support earlier research on Cr (VI) adsorption using various

adsorbents that follow pseudo-second-order kinetics (Badessa et al., 2020; Garg et al., 2023).

3.5 Adsorption capacity comparison of various agricultural waste adsorbents

Table 2 compares the current study's adsorption capacity (q_m) for Cr (VI) ions to those recently reported by other researchers utilizing different adsorbents without chemical treatment or modifications applied. Apparently, banana pseudo stem adsorbent has a better adsorption capacity than some of these adsorbents while being comparable to others. The different adsorption capacities of these adsorbents can be attributed to a variety of factors.

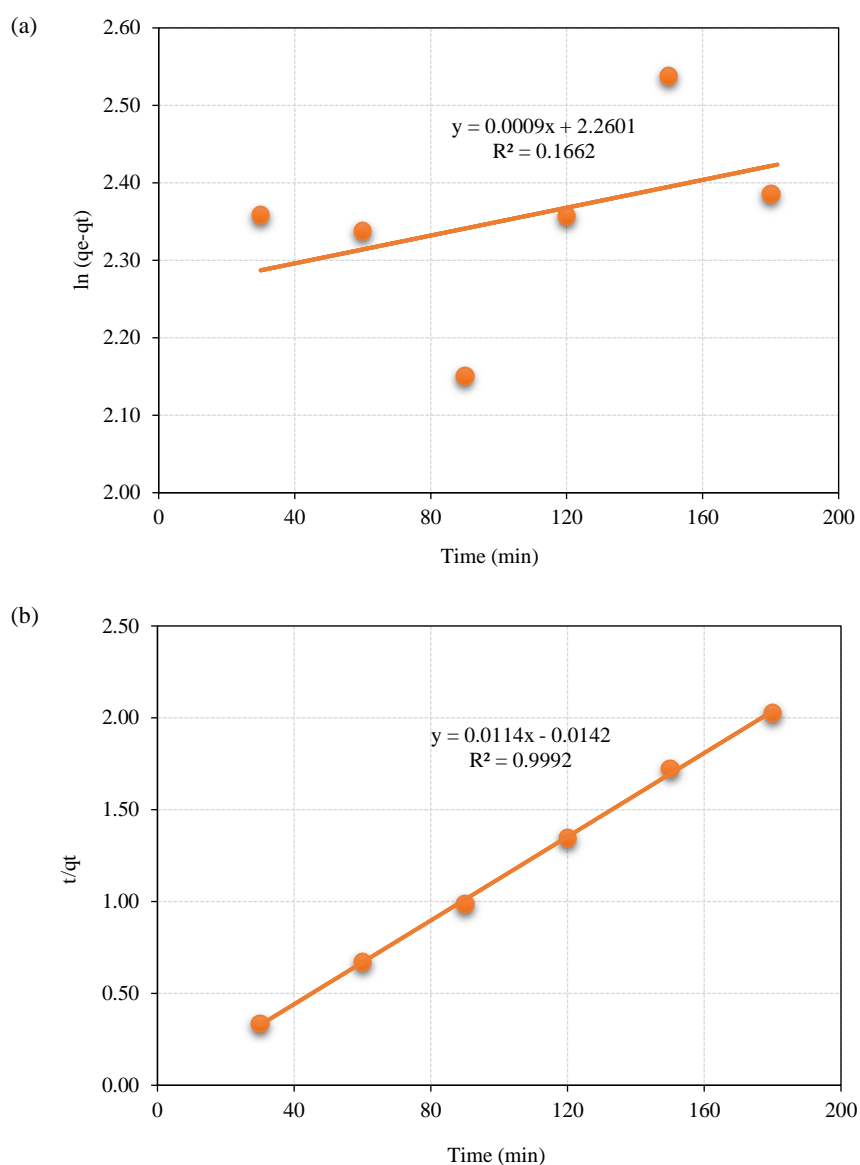


Figure 7. Kinetic of Cr (VI) ions adsorption by, (a) pseudo-first-order; (b) pseudo-second-order models

Table 2. Comparative analysis of the Cr (VI) adsorption capability of several adsorbents

Types of agriculture waste adsorbent	Adsorption capacity, q_m (mg/g)	References
Canary banana peels	10.09	Martín et al. (2016)
Palm fiber	6.00	Abubeah et al. (2018)
Groundnut shell	3.79	Bayuo et al. (2019)
Cranberry kernel shell	6.81	Parlayici and Pehlivan (2019)
Corn cob	0.54	Melese et al. (2020)
Wheat straw and <i>E. adenophorum</i>	89.22	Song et al. (2021)
Sugarcane bagasse	1.49	Abilio et al. (2021)
Banana pseudo stem	33.33	Current study

4. CONCLUSION

In this study, banana pseudo stem adsorbent showed great potential for removing Cr (VI) ions from an aqueous solution. From the batch adsorption experiments, the produced adsorbent successfully

removed the Cr (VI) ions up to 91.9% with the following optimal adsorption parameters, pH 2, the dosage of 0.5 g, initial Cr (VI) concentration of 500 ppm and 90 min contact time. The removal percentage increases with declining pH, rising adsorbent dosage

and initial concentration, and longer contact times. The Langmuir isotherm model better fit the equilibrium experimental results, with a maximum monolayer adsorption capacity (q_m) of 33.33 mg/g. The adsorption kinetics was best described by the pseudo-second-order kinetic model, indicating the physical adsorption. In conclusion, banana pseudo stem wastes offer an attractive, eco-friendly, simple, and economically alternative adsorbent for treating and removing Cr (VI) ions from wastewater. It is recommended to convert banana pseudo stem as biochar and compare its adsorption and Cr (VI) ions removal potential to this current study. In keeping with government policy and the Sustainable Development Goals (SDG), it is expected that this study would aid the related industries to reduce the negative effects of banana plant waste and chromium contamination on the environment, public health, economy, and society.

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