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Adsorption of Chromium Using Apu Wood (*Pistia stratiotes L*) Root Bioadsorbent and Modification of Apu Wood Root

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ABSTRACT

The adsorbent is a material used for the adsorption process. One alternative used in the selection of adsorbents is biological materials. One type of plant that can be used to adsorb heavy metals is Apu Wood. This study aims to examine the effect of Cr (VI) concentration on the adsorption capacity of adsorbent, examine the time adjustment to the adsorption capacity of adsorbent, examine the characterization of adsorbent using SEM and FTIR, assess the adsorption equilibrium using the Langmuir and Freundlich methods and the kinetics of the adsorption reaction using pseudo order 1 and pseudo order 2. In this study, variables are the concentration of initial solution Cr(VI) of 2, 4, 6, 8, and 10 ppm and contact time of 15, 30, 45, 60, and 75 minutes. The result of this research is the capacity of adsorption increases in line with the increase in the initial concentration of Cr(VI) solution as adsorbate. Contact time affects the capacity of adsorption where the longer the contact time, the adsorption capacity increases until it reaches equilibrium at 70 minutes. In SEM and FTIR analysis both tests showed that aluminum adsorption using apu wood adsorbent and its modification changed the composition of the function group and the structure of the adsorbent surface area. The Langmuir Isotherm model produces a greater value R2 than the Freundlich isotherm model which indicates that the adsorbent surface is in direct contact with adsorbate and adsorbate attaches to the active site of the adsorbent of only one layer (monolayer). Kinetics adsorption pseudo order 1 produces a value of R2 greater than kinetics adsorps pseudo order 2 which indicates the mechanism that occurs is the transfer of mass in addition to the use of electrons together between heavy metals Cr (VI) with apu wood root.

1. Introduction

Industrial processes such as mining, electroplating, or battery manufacturing generate heavy metal waste that is dumped into the aquatic ecosystem. Heavy metals are toxic pollutants that can accumulate and have a negative impact on the environment. Removing toxic contaminants from wastewater is one of the most important things for the environment. Heavy metals contained in wastewater can damage aquatic ecosystems. Even at low concentrations, the effects of heavy metal ions can have a direct effect until they accumulate in the food chain. Heavy metals in waste usually exist in various conditions such as insoluble, dissolved, reduced, oxidized, and complex. Heavy metals that cannot be decomposed must be removed for better environmental quality [1].

There are several methods that have been developed to remove heavy metals, ranging from extraction, ion exchange, chemical precipitation, separation using membranes, and adsorption [2]. The method that has been developed has several weaknesses, namely high installation and operational costs require a lot of chemicals and produces waste that requires further processing in its disposal. Of all the methods that have been developed, adsorption is the simplest method and the easiest to operate. Adsorption is a process that occurs when a gas or liquid accumulates on the surface of a solid or liquid object called an adsorbent.

The adsorbent is a material used for adsorption where the process occurs on the surface [2]. Adsorbents are categorized into three types: 1. Oxygen-containing materials are usually polar and hydrophilic (silica gel and zeolite); 2. Materials consisting of carbon arrays are usually nonpolar and hydrophobic (activated carbon and graphite); 3. The material of the polymeric arrangement is usually polar or nonpolar with functional groups in a porous polymer matrix [3]. In addition, adsorbents must have high adsorption capacity, are relatively inexpensive, and can be obtained easily [4].

One alternative used in the selection of adsorbents is the use of biological materials as adsorbents. This process is referred to as biosorption. Biosorption is a waste treatment that can remove heavy metals in liquid waste by biological materials. This biological material is used because it is relatively cheap, can be obtained easily, and is environmentally friendly. This biosorption process utilizes the cellulose content in plants. Cellulose is a compound that has a hydrophilic character due to the presence of a hydroxyl group in each polymer unit [5]. The surface of the natural cellulose functional groups and their derivatives can interact physically or chemically. Cellulose has functional groups that can bind with metal ions. The functional group is the hydroxyl group. One type of plant that can be used to adsorb heavy metals is apu wood.

Apu wood (*Pistia stratiotes L.*) is a plant that can be found floating in calm waters or ponds. Apu wood is one of the phytoremediator plants, namely plants that have the ability to treat waste in the form of heavy metals, and organic and inorganic substances. This plant has a high fiber content. Mishima et al. [6] stated that the roots of apu wood contain 16.5% cellulose. This high fiber content is expected to be used as a source of cellulose as a new alternative for heavy metal adsorbents.

In this study, adsorption of heavy metal chromium (VI) was carried out using the roots of apu wood after it was activated and the roots of apu wood that had been modified by converting it into activated carbon. The adsorbent from the roots of apu wood was carried out because of its abundant presence, high content of cellulose and fiber, and easy preparation process so the cost used was relatively low.

2. Research Method

2.1 Materials and Tools

2.1.1 Material

The material used is root of apu wood from the UNDIP reservoir in the form of fibrous roots with leaf sizes of 5-14 cm. Technical $K_2Cr_2O_7$, Aquadest, NaOH 1% (v/v), H_2O_2 1% (v/v), technical NaCl.

2.1.2 Tools

The tools used are oven, miller, screen, universal pH, magnetic stirrer, Whatman no 42 filter paper, Furnace, Atomic Absorption Spectrophotometry (AAS), FTIR (Fourier Transform Infrared), and SEM (Scanning Electron Microscopy).

2.2 Method

2.2.1 Preparation of 100 ppm. Chromium (VI) Heavy Metal Solution

Potassium dichromate ($K_2Cr_2O_7$) was weighed as much as 0.2827 grams and then put into a 1000 ml volumetric flask. Potassium dichromate was dissolved with aquademin to the mark and then the chromium (VI) solution was tested using AAS.

2.2.2 Adsorbent Preparation

The roots of the grasshoppers were separated from the leaves and washed with running water 3-5 times. The roots of the grasshoppers were then dried in the sun for 7 days and then placed in an oven at 100 °C for 24 hours. Then the dried grasshopper roots were crushed using a miller. Apu wood root powder was soaked with 1% (w/v) NaOH for 24 hours. Then the roots of the apu wood were added with 1% (v/v) H_2O_2 and soaked for 24 hours. The mixture was filtered and rinsed with distilled water to pH 6-8. The filter results were dried in an oven at 100 °C for 24 hours. The dried grasshopper root powder was sieved using a 100 mesh sieve [6].

2.2.3 Modification of Apu Wood Root

The roots of apu wood are separated from the leaves and rinsed with running water 3-5 times. The roots of apu wood were dried in the sun for 7 days and then placed in an oven at 105 °C. The dried apu wood roots were soaked in NaCl with a mass ratio of 1:1. After that, the roots of apu wood are dried. The dried wood roots were carbonated at 200 °C for 2 hours. The carbonation results were rinsed several times using distilled water. After that, it was dried, then sieved at a size of 100 mesh.

2.3 Adsorption

2.3.1 Effect of Time on Adsorption Capacity

Apu wood root powder and apu wood root modification were weighed as much as 1 gram each and then added to the metal solution. The solution was stirred with a magnetic stirrer for 15, 30, 45, 60, and 75 minutes at room temperature with a stirring speed of 150 rpm and then filtered through filter paper. Each filtrate was measured using AAS. Pulp powder and modification of the roots of pumice were analyzed using SEM and FTIR.

2.3.2 Effect of Heavy Metal Concentration on Adsorption Capacity

Cr(VI) solutions with concentrations of 2, 4, 6, 8, and 10 ppm were prepared. Apu wood root powder and apu wood root carbon were weighed as much as 1 gram each and then added to the prepared Cr (VI) metal solution. The solution was stirred with a magnetic stirrer at room temperature with a stirring speed of 150 rpm and then filtered using filter paper. Each filtrate was measured using AAS. Pulp powder and root carbon of pumice were analyzed using SEM and FTIR.

2.3.3 Characterization Test Using SEM

The sample is placed and attached to the SEM specimen holder using a carbon double type with the cross-section pointing vertically upwards or an objective lens. This double tip is made of conductive carbon material on both sides which functions to deliver all electrons that enter the sample out through grounding. The sample chamber is vacuumed to 10-6 torr to ensure that the SEM column is free of air molecules. SEM is operated with standard operating parameters of High Voltage of 20 kV, Spot Size of 50, and Work Distance (WD) of 10 mm [7].

2.3.4 Characterization Test Using FTIR

Samples in the form of powder are placed on the preparation and pressed with a press to form pellets. Next, the pellet is placed in the sample holder. The sample holder was inserted into the preheated instrument for 15 minutes, then the lamp was turned on right on the sample, with the wavenumber set at 400-4000 cm^{-1} [8].

3. Result and Discussion

3.1. Effect of Initial Concentration of Cr (VI) on the Adsorption Capacity of Apu Wood Roots and Modified of Apu Wood Roots

In this study, the effect of the initial concentration of Cr (VI) on the adsorption capacity of apu wood roots and modification of apu wood roots was studied at a temperature of 25 °C at the initial concentrations of 2, 4, 6, 8, and 10 ppm. The research results can be shown in Fig 1.

The value of qe or adsorption capacity is the amount of heavy metal that can be adsorbed (mg) in 1 gram of adsorbent. The results showed that the initial concentration affected the efficiency of Cr metal adsorption. As seen in Figure 1 the adsorption capacity of the roots of apu woods obtained from a concentration of 2 ppm to 10 ppm in a row is 1.001; 1.567; 1.780; 2.184; and 2.481 mg/g while for modified apu wood roots it was 1.002; 1.550; 1.978; 2.283; 2.583 mg/g. The adsorption capacity increased along with the increase in the initial concentration of Cr(VI) solution as adsorbate. This is due to the availability of the active site of the adsorbent which is still able to absorb more Cr (VI) ions in solution for adsorption [9].

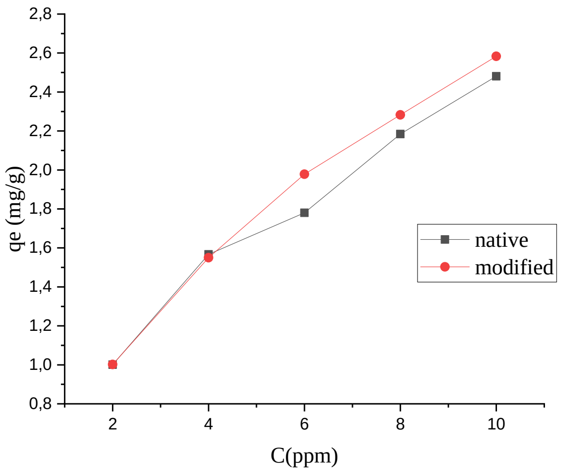
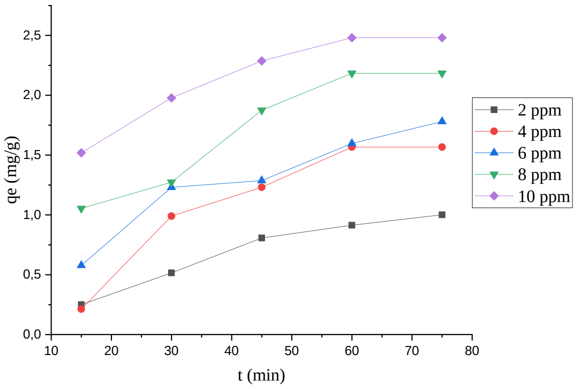


Fig. 1. Effect of initial concentration on the adsorption capacity of Apu wood roots and modification of Apu wood roots

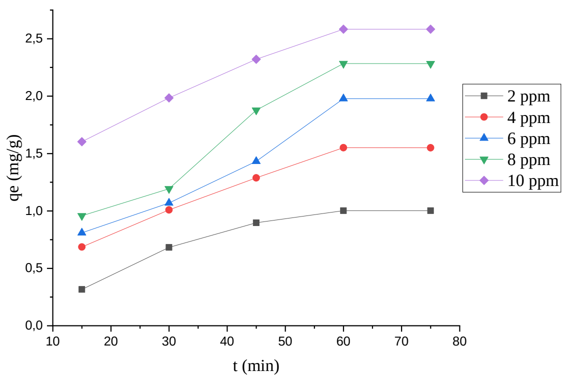
In Fig 1, the adsorption capacity increases with the increase in the initial concentration of Cr (VI) solution as adsorbate. However, there was a difference where the adsorption capacity of the modified Apu root was higher than that of the root. This is because modifications to the roots of apu wood cause an increase in the surface area of the adsorbent, an increase in affinity with Cr (VI), and an increase in adsorption capacity [10].

3.2.The Effect of Time on the Adsorption Capacity of Apu Wood Roots and Modified of Apu Wood Roots

The effect of time on the adsorption capacity of the Apu wood roots and the modification of the Apu wood roots was investigated at 25 °C at contact times of 15 minutes, 30 minutes, 45 minutes, 60 minutes, and 75 minutes as shown in Fig 2.



(a)



(b)

Fig. 2. Effect of time on qe of Apu wood roots (a) and modification of apu wood roots (b)

The results showed that time had an effect on the adsorption capacity of apu wood roots and the modification of apu wood roots. From Fig 2 it can be seen that the length of contact time results in a higher of adsorption capacity. The longer of the adsorption process was carried out lead to the higher contact between the adsorbent and the adsorbate. This is because there is still an active group capable of absorbing the adsorbate [11]. At the time of 60 minutes and 75 minutes, the increase in adsorption capacity was not significant or tended to be constant. This is because the active group has adsorbed most of the adsorbate and indicates that the adsorbent is already saturated [12].

In Fig 2, there is a difference in the adsorption capacity of Apu wood root (a) and modification of Apu wood root (b). The adsorption capacity of modified Apu wood root was greater than that of 1.04%. This is because modifications to the Apu wood roots cause an increase in the surface area of the adsorbent, an increase in affinity with Cr (VI), and an increase in adsorption capacity [10].

3.3. Study of Characterization of Apu Wood Roots and Modifications of Apu Wood Roots

3.3.1. Analysis of Functional Groups of Apu Wood Roots and Modification of Apu Wood Roots

Analysis of the characteristics of the functional groups contained in the Apu wood roots using the analytical method shown in Fig 3. In this case, information was obtained about the composition of the functional groups indicated by the presence of a peak at a certain wavenumber [13].

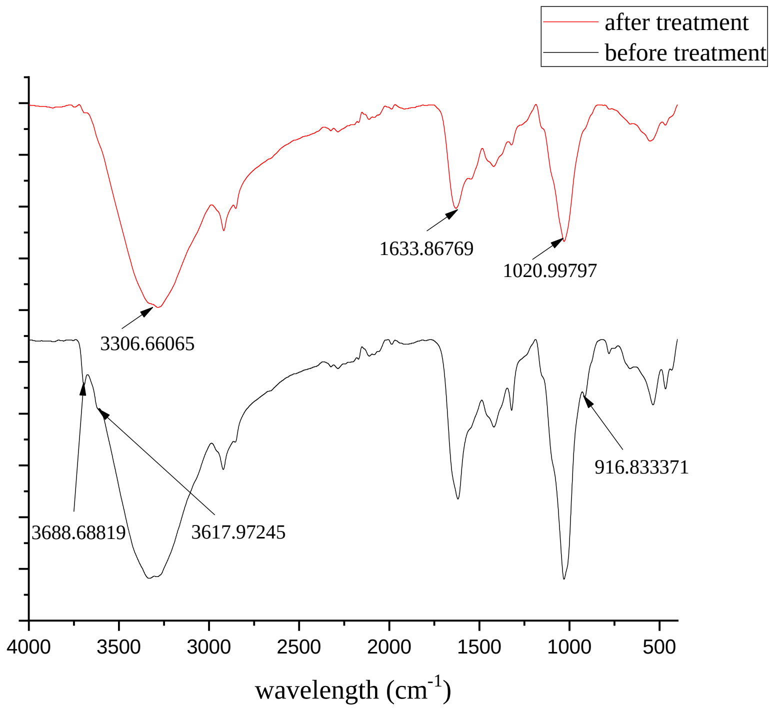


Fig. 3. Results of FTIR analysis of Apu wood root adsorbents (native) before and after treatment

Comparison between Apu wood root adsorbents (native) before and after treatment with Cr (VI). There are several peaks that indicate functional groups in the adsorbent. Peak 3688.68819 – 3306.66065 cm^{-1} indicates the presence of an O-H stretching group which is a hydroxyl group with an adsorbance range of 3550-3200 cm^{-1}. At peaks of 3688.68819 cm^{-1} and 3617.97245 cm^{-1} were groups that were lost after treatment with Cr (VI) caused by the adsorption of heavy metals by hydroxyl groups [13]. At peaks of 1633.86789 cm^{-1} and 1613.54708 cm^{-1}, it shows the presence of a bending N-H functional group which is an amine group in the adsorbance range of 1650-1580 cm^{-1}. At the peak of 1021.67043 cm^{-1} and 1020.99977 cm^{-1}, it shows the presence of a C-N stretching group which is an amine functional group. According to Vieira [13], the functional groups that play an important role in the absorption of heavy metals are amines, carboxylates, hydroxyl, sulfonates, and thiols.

Fig 4 shows the comparison between the modified Apu wood root charcoal adsorbent before and after treatment with Cr (VI). There are several peaks that indicate the functional groups in the adsorbent. At the peak of 3333.48386 cm^{-1} and 3332.0000903 cm^{-1}, it shows the presence of an O-H stretching group which is a hydroxyl group in the adsorbance range of 3550-3200 cm^{-1}. The peaks of 1607.58465 cm^{-1} and 1605.58465 cm^{-1} indicate the presence of a bending N-H group which is an amine group in the adsorbance range of 1650-1580 cm^{-1}. At the peak of 1103.90607 cm^{-1}, it shows the C-O stretching group (secondary alcohol) which after contact with Cr (VI) the group disappears because chromium is a strong oxidizing agent so chromium oxidizes the group [14]. At the peak of 1036.4416 cm^{-1} and 1034.67269 cm^{-1} it shows the S=O (sulfoxide) group in the adsorbance range of 1070-1030 cm^{-1}. According to Vieira [13], functional groups that play an important role in the absorption of heavy metals are amines, carboxylates, hydroxyls, sulfonates, and thiols.

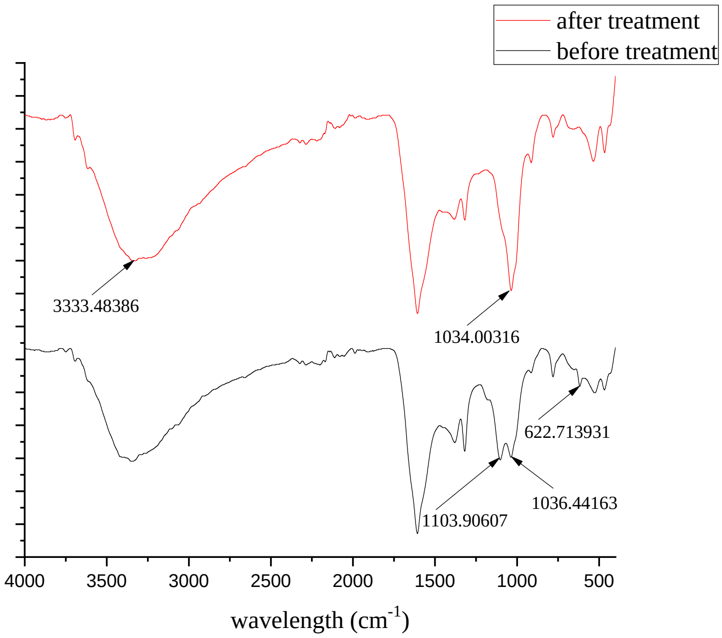
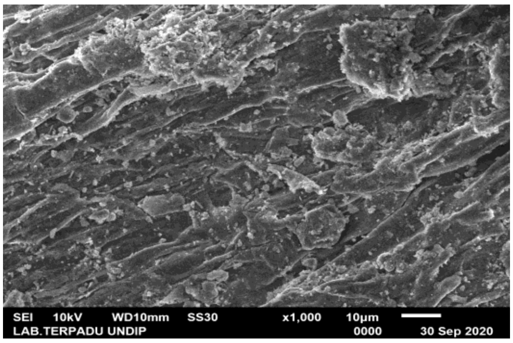


Fig. 4. The results of the FTIR analysis of Apu wood root charcoal (modified) before and after treatment

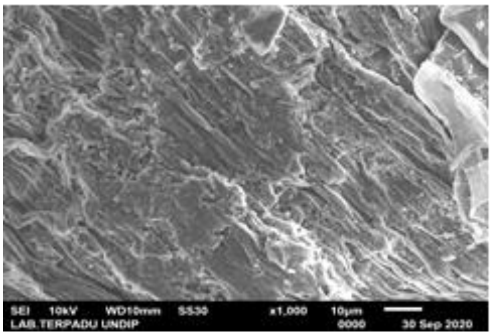
In this study, the adsorbents of the native and modified root of the apu wood have groups that are important in the absorption of heavy metals.

3.3.2. Morphology Analysis of Apu Wood Root Surfaces and Modification of Apu Wood Roots

The surface morphology of the apu wood was tested using SEM (Scanning Electron Microscope) with 1000x magnification. The results of the SEM analysis of pumice roots and modifications of pumice roots can be seen in Fig 5.



(a)



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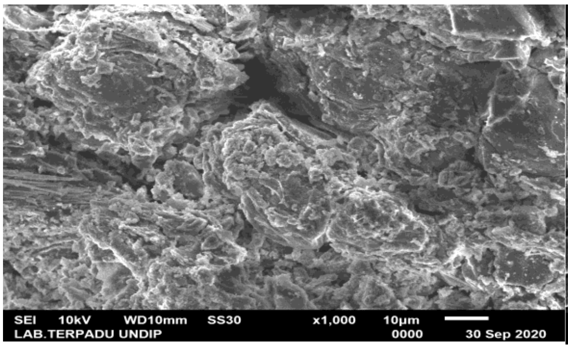

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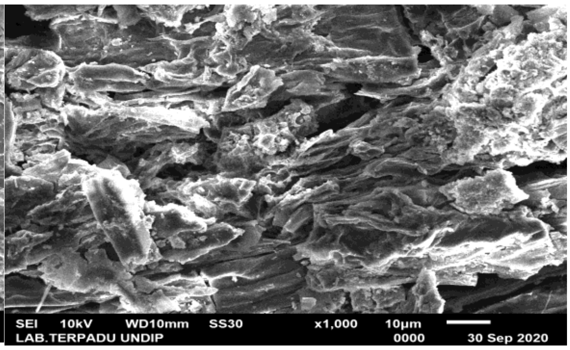

(d)

Fig. 5. SEM analysis results a) native before, b) after treatment, c) modified before, d) after treatment

As shown in Fig 5 the difference in surface area between the native sample before treatment and the native sample after treatment. It can be seen that after the chromium adsorption treatment, a rougher surface is formed than the raw native. As for the modified sample, the difference in surface area can be observed that after the sample is treated with chromium the surface area becomes rougher and has an irregular shape. This is due to the adsorption of the adsorbent on the chromium.

3.4. Study of the Data on the Adsorption Isotherm Equilibrium

In this study, the adsorption isotherm model to be compared for Cr(VI) adsorption are Langmuir and Freundlich. The adsorption isotherm model is an explanation of the interaction between the adsorbate and the adsorbent. Langmuir adsorption isotherm is a model that explains that the surface of the adsorbent is in direct contact with the adsorbate and the adsorbate attaches to the active site of the adsorbent only in one layer/monolayer [3]. While the Freundlich adsorption isotherm is a model that explains that the surface of the adsorbent is heterogeneous with various active sites and different energies. The results of the study can be seen in Fig 6.

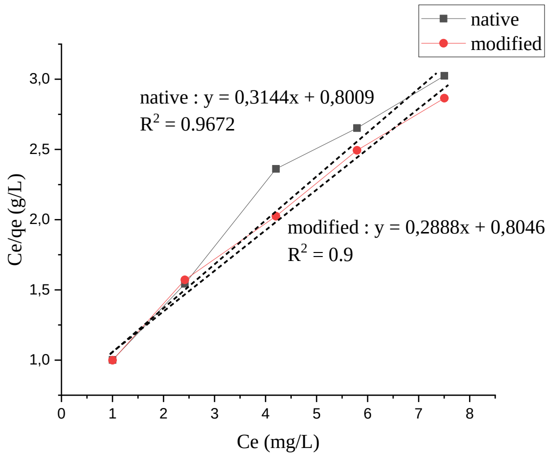

Fig. 6. Langmuir adsorption isotherm of Apu wood root and modification of Apu wood roots

Based on Fig 6, the equation y = 0.3144x – 0.8009 and an R^2 value of 0.962 for the adsorption of the Apu wood root, while for the modification of the Apu wood roots, the equation y = 0.2888x - 0.8046 and the R^2 value of 0, 9781. From this equation, the slope and intercept values are obtained which are used to find the values of Q_m and K_L. In this case, it is the maximum adsorption capacity of the monolayer. While the Freundlich adsorption isotherm is a model that explains that the surface of the adsorbent is heterogeneous with various active sites and different energies. Freundlich adsorption isotherm graph can be seen in Fig 7.

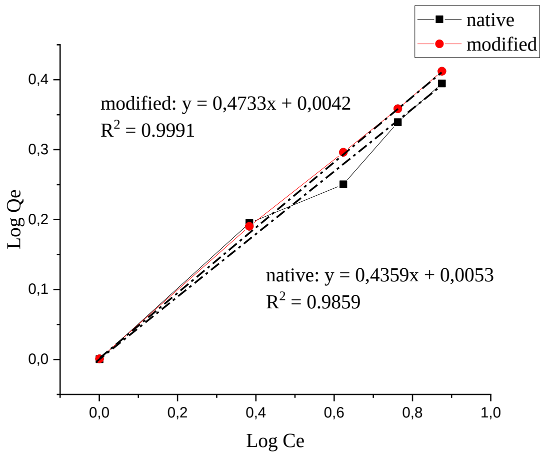

Fig. 7. Freundlich adsorption isotherm of Apu wood root and modification of Apu wood root

Based on Fig 7, the equation y = 0.43659x – 0.0053 and the R^2 value of 0.9859 for the adsorption of the roots of the apu wood root is obtained. For the modification of the apu wood root, the equation y = 0.4733x – 0.0042 and the R^2 value of 0.9991. From this equation, the slope and intercept values are obtained which are used to find the values of kf and n. In this case, kf is an isothermal constant, while n is the adsorption intensity.

Table 1. Model of Adsorption Isotherm of Apu wood root and Modification of Apu wood root

Isotherm Model	Adsorbent	Parameters	R^2
Langmuir Equation	Apu Wood Root	KL = 0.3569	0.9672
		Qm = 3.1807	
	Modified Apu Wood Root	KL = 0.3600	0.9000
		Qm = 3.4626	
Freundlich Equation	Apu Wood Root	Kf = 1.0122	0.9859
		1/n = 0.4359	
	Modified Apu Wood Root	Kf = 1.0143	0.9991
		1/n = 0.4733	

As shown in Table 1 on the Langmuir adsorption isotherm, the Q_m values for the adsorption of apu wood and modified apu wood were 3.1807 and 3.4626 mg/g, respectively. This means that 1 gram of apu wood is able to adsorb 3.1807 mg of Cr (VI) metal and modified of apu wood is able to adsorb 3.4626 mg of Cr (VI) metal. With this, it can be concluded that the adsorption capacity of the modified roots of the apu wood is greater than the Apu wood root.

The isothermal constants obtained from the Freundlich isotherm equation which become the parameters for adsorption capacity are Kf and 1/n. As shown in Table 1, the Kf and 1/n values for adsorption with apu wood roots were 1.0122 and 0.4359, respectively. The Kf and 1/n values for adsorption with modified apu wood roots were 1.0143 and 0.4733, respectively. These two constants are very important to choose an adsorbent as a separating medium. Kf is the overall adsorption capacity and 1/n is the heterogeneity factor that indicates the strength of the bond energy between the adsorbate and adsorbent particles. In the adsorption process, the higher value of Kf and 1/n result in more economically feasible the adsorbent is [15].

From these two adsorption isotherm models, it can be seen that the Langmuir isotherm has a higher R^2 value than the Freundlich isotherm. This shows that the surface of the adsorbent is in direct contact with the adsorbate and the adsorbate attaches to the active site of the adsorbent only in one layer (monolayer). The value of R^2 which is 0.9 and close to 1 gives a good agreement with the research and indicates favorable adsorption conditions. Langmuir and Freudlich adsorption isotherms can be applied to the adsorption of Cr (VI) with the roots of apu wood.

3.5. Study on the Kinetics of Adsorption using Pseudo 1st Order and Pseudo 2nd Order

The kinetics of the adsorption reaction was carried out to find out about the adsorption mechanism or controlling rate, such as mass transfer or the chemical reaction process of heavy metal Cr (VI) in the Apu wood roots and modification of Apu wood roots as shown in Fig 8.

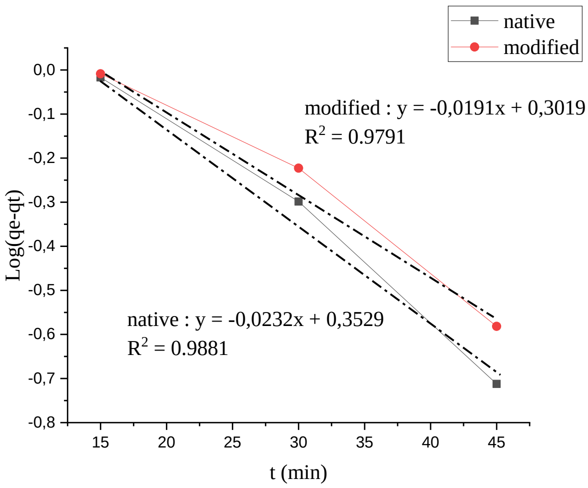

Fig. 8. Reaction kinetics of Pseudo 1st Order adsorption of Apu wood root and modification of Apu wood root

The pseudo 1st order equation can describe the adsorption mechanism that occurs. The pseudo 1st order equation illustrates that the mechanism that occurs is an alternating reaction between the adsorbent and the adsorbate at equilibrium. It also means that mass transfer acts as a rate limiting step in the adsorption process

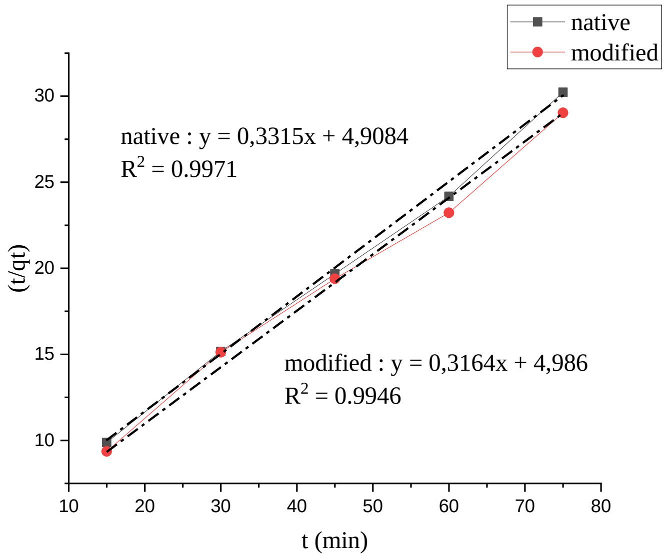

Fig. 9. Reaction kinetics of Pseudo 2nd Order adsorption of Apu wood root and modified Apu wood root

Table 2. Adsorption kinetics of Pseudo 1st Order and 2nd Order

		Apu Wood Root	Modified Apu Wood Root
Pseudo 1st Orde	qe	1.7778	2.004
	k1	0.0497	0.044
	R^2	0.9881	0.9791
Pseudo 2nd Orde	qe	0.3454	2.2537
	k2	0.0299	0.0224
	R^2	0.9971	0.9946

Table 2 shows the value of R^2 and the reaction rate constant of the two adsorption kinetics models used in this study. Judging from the value of R^2 adsorption kinetics for apu wood and modification of apu wood root gives a better tendency in the pseudo 2nd order equation. This supports that the mechanism that occurs is mass transfer in addition to the sharing of electrons between the heavy metal Cr (VI) with the Apu wood roots and modification of the Apu wood roots. This can happen because there are functional groups such as hydroxyl which will bind metal ions through chemical and physical adsorption mechanisms [16].

4. Conclusion

Research on chromium adsorption using Apu wood root bioadsorbent and modification of apu wood root resulted in several conclusions, namely the adsorption capacity increased with the increase in the initial concentration of Cr (VI) solution, contact time affected the adsorption capacity where the longer of the contact time results in the adsorption capacity increased until it reached equilibrium. at 70 minutes, SEM and FTIR analysis showed that changing the arrangement of functional groups and the structure of the surface area of the adsorbent, the Langmuir isotherm model produced a higher R^2 value than the Freundlich isotherm model, pseudo-adsorption kinetics of 1st order resulted in a higher R^2 value than the pseudo 2nd order which shows the mechanism that occurs is mass transfer in addition to the sharing of electrons between heavy metal Cr (VI) with apu wood roots and modification of apu wood roots.

Notation

C	= initial concentration of Cr(VI), ppm
qe	= adsorption capacity, mg/g
qt	= adsorption capacity at time t, mg/g
Ce	= concentration at equilibrium, mg/L
Qm	= maximum adsorption capacity, mg/g
KL	= Langmuir's constant
Kf	= Freundlich constant
1/n	= heterogeneity factor
k1	= pseudo-order rate constant of reaction 1
k2	= pseudo-order rate constant of reaction 2

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