Langmuir and Freundlich isotherm equation test on the adsorption process of Cu (II) metal ions by cassava peel waste (*Manihot esculenta crantz*)

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Abstract. This Study aims to determine the adsorption process of Cu (II) metal by cassava peel waste (*Manihot esculenta crantz*) meeting the Langmuir equation and Freundlich. Research method cassava peel made into activated carbon, then determine the mass, pH and optimum contact time. Then the determination of the maximum adsorption capacity was carried out by testing with the Langmuir isotherm and Freundlich isotherm equations. The results showed that the best conditions (optimal conditions) were obtained with the addition of 0.5 grams of active carb from cassava peels. The percentage of Cu ion adsorption in these conditions was 97.72%, at pH 6 and a contact time of 60 minutes and the absorption capacity was 98.49%. The maximum adsorption capacity of cassava peel activated carbon to Cu(II) ions at the optimum condition was determined based on the Langmuir and Freundlich isotherm equations. The results obtained were -51.813 mg/g and 26,792 mg/g, respectively.

1. Introduction

Along with the times and accompanied by population growth and industrial development, it triggers an increase in the amount of industrial waste that can cause environmental pollution. One of them is water pollution by waste containing heavy metals, such as Cu. The impact of being contaminated with Cu metal in the body causes stomach cramps, nausea, vomiting, diarrhea, kidney failure, liver and even death [1]. Copper metal (Cu) is found in water and soil both in the form of ions and compounds. Naturally Cu enters the waters through erosion, rock erosion, or in the atmosphere brought down by rainwater [2]. Although heavy metal Cu (II) is an essential element for the body, it is in an amount that exceeds the threshold because with industrial developments it can be harmful to health.

Along with the development of the industry, various efforts were made to reduce pollution caused by these heavy metals. One of the methods that have been developed to remove heavy metals is adsorption [3]. Adsorption is the process of absorption by certain solids against certain substances that takes place on the surface of the solid due to the attraction of atoms or molecules on the surface of the solid without seeping into it. The solid surface is referred to as the adsorbent, whereas the adsorbed substance is referred to as the adsorbate [4]. The adsorbent which is generally used is activated carbon. Activated carbon is an amorphous compound derived from materials containing carbon or charcoal which are specially treated to obtain high absorption energy [5].

The adsorption process is currently being developed by utilizing waste containing carbon, including banana peels, coffee peels, rice husks, cocoa peels, durian peels. Besides this waste, cassava peel is also a material that contains carbon and has the potential as an adsorbent. Currently, this waste has not
been utilized optimally, while the potential availability of cassava peels is very abundant. Based on survey data, cassava peel waste in the city of Palu, Central Sulawesi, reached 147 kg/per day. One of the producers of cassava peel waste which is quite large is the cassava chip manufacturing industry.

The superiority of cassava peel compared to durian peel waste, banana peel, cocoa peel and coffee peel as an adsorbent because it contains lignocellulose which has the potential to adsorb cationic waste. Lignocellulose is a natural material consisting of hemicellulose, cellulose and lignin. The main functional groups on the surface of lignocellulosic materials are hydroxyl, aldehyde, carboxylate, and groups Cyano [6]. Therefore, materials containing lignocellulose, such as cassava peel, become an alternative as an adsorbent that can be utilized, because of its large availability and valuable because it is only used as waste.

Research data from cassava peel contains carbon (C) of 59.31%, hydrogen (H) of 9.78%, oxygen (O) of 28.74%, nitrogen (N) of 0.11%, and water (H₂O) by 11.4% [7]. Based on the high carbon content, cassava peel has the potential to be used as raw material for activated carbon with an activation and carbonization process that can be used as an adsorbent [8].

Activated carbon from cassava bark has been used for testing the absorption of Fe ions and obtained the highest absorption of 51.97% at the carbonization temperature of 600°C [9]. The same study used activated carbon from corn cobs as Cu (II) ion adsorbent. The results showed that the adsorption of Cu (II) metal occurred at an optimum pH of 7 with an absorption of 98.34%, the optimum contact time occurred at 60 minutes with the percentage of Cu (II) adsorbed at 96.37%, the maximum adsorption capacity of Cu (II) ions of 2.4160 mg/gr [10]. Based on the results of the research on the adsorption of cassava peel activated carbon to Fe (II) ions and corn cobs activated carbon adsorption to Cu(II) ions, in this study, the optimum pH determination was used variations of 4-9, the mass of active carbohydrates was 0.3-0.6 gram and variation of contact time 15, 30, 60 and 90 minutes. These variations were used to absorb Cu(II) ions using activated carbon from cassava peels, while for testing the absorption capacity in this study the Langmuir and Freundlich isotherm equations were used.

2. Methods
The materials used in this study were cassava peels obtained from local traditional markets. Other chemicals such as 0.1 N NaOH solution, 0.1 N HCl solution, solid CuSO₄.5H₂O, were obtained from E. Merck and used without further purification.

2.1. Sample Preparation
The Cassava peel was washed thoroughly and separated from the brown outer skin then cut into small pieces and then dried in the sun for 2 days [11].

2.2. Making Adsorbent
Dry cassava peels are put into a furnace to make charcoal at 400°C for 1 hour and then allowed to cool at room temperature. The cassava peel charcoal was mashed using a blender until the charcoal passed through a 100 mesh sieve. Cassava peel drying and carbonation in this process aims to decompose hydrocarbon compounds such as cellulose and hemicellulose into pure carbon. Carbonated charcoal was ground and sieved using a 100 mesh sieve. With the purification process, the chemical activation process will be more evenly distributed because the smaller the size of the cassava peel carbonation charcoal, the greater the surface area that will be in contact with the activator so that more activated carbon and more pores are formed on each carbon. particles [12].

2.3. The Activation Process of Cassava Peel
The activation process is carried out by chemical means, namely cassava peel charcoal soaked in 0.1 N NaOH solution for 60 minutes. Then the activated carbon produced was washed with aquadest and 0.1 N HCl until the filtrate had a neutral pH (pH 6 to 7) which was measured using a pH meter. Then filtered using filter paper. Samples obtained dried in an oven at 1100C for 60 minutes [12]. Activation of cassava peel charcoal aims to increase metal absorption, because NaOH is very hygroscopic (easy to bind water) so that the H₂O contained in the charcoal can react with it [11].
2.4. Preparation of Stock Solution of Cu (II) 1000 ppm
Copper Manufacture mother liquor with a concentration of 1000 ppm is by weighing the solids \( \text{CuSO}_4 \cdot 5\text{H}_2\text{O} \) as much as 3.929 gram using analytical balance and then put into the flask, then add 1000 mL of aquadest up to the mark limit, then shaken until the solid dissolves and is ready to use.

2.5. Determination of optimum mass
The adsorbent mass (cassava peel activated charcoal) used was 300 mg, 400 mg, 500 mg, 600 mg, and 700 mg. Each was put into an Erlenmeyer then added 50 mL of 50 ppm Cu(II) solution, then stirred for 60 minutes using a shaker, then filtered using filter paper, then the adsorbance was measured using an atomic absorption spectrometer [13].

2.6. Determination of the optimum solution pH
50 mL of Cu (II) solution was added to each Erlenmeyer with a concentration of 50 ppm, then the optimum mass of cassava peel adsorbent was added to the Erlenmeyer. Then each Erlenmeyer pH was adjusted with variations of 5, 6, 7, and 9, then the mixture was stirred for 60 minutes using a shaker, then filtered using filter paper and the adsorbance was measured using an atomic absorption spectrometer [13].

2.7. Determination of optimum contact time
50 mL of Cu (II) solution was added to each Erlenmeyer with a concentration of 50 ppm, then the optimum mass of cassava peel adsorbent was added to the Erlenmeyer and adjusted the pH of the solution at the optimum pH (treatment result f). Then each Erlenmeyer was set the stirring time with variations of 15, 30, 60, and 90 minutes, then stirred using a shaker, then filtered using filter paper and then the absorbance was measured using an atomic absorption spectrometer [13].

2.8. Determination of adsorption capacity with variations in concentration
Cu(II) mother liquor is added to each Erlenmeyer with a concentration of 150, 250, and 300 ppm as much as 50 mL, then the optimum mass of cassava peel adsorbent is added to each Erlenmeyer by setting it at the optimum pH, then stirred using a shaker during the optimum contact time obtained, then filtered using filter paper, then measured the adsorbance using an atomic absorption spectrometer. The Capacity (capacity adsorption) of the cassava peel waste adsorbent was determined by the Langmuir isotherm and the Freundlich isotherm [13].

2.9. Data analysis technique
The data analysis used in this study is the Langmuir isotherm (equation 1) and the Freundlich isotherm (equation 2)

\[
\frac{C}{x/m} = \frac{1}{\alpha \beta} + \frac{1}{\alpha} C \tag{1}
\]

Information:
\( \frac{x}{m} = \) adsorption power of the amount of adsorbate adsorbed per unit mass of adsorbent (mg/g)
\( C = \) equilibrium concentration of adsorbate in solution after adsorption (ppm)
\( \alpha = \) maximum adsorption power
\( \beta = \) Langmuir constant [14].

\[
\log \frac{x}{m} = \log k + \frac{1}{n} \log C \tag{2}
\]

Information:
\( \frac{x}{m} = \) adsorption power of adsorbate amount adsorbed per unit mass of adsorbent (mg/g)
\( C = \) equilibrium concentration of adsorbate in solution after adsorption (ppm)
\( n = \) Freundlich’s constant
\( k = \) power maximum adsorption (mg/g) [14].
3. Results and Discussion

3.1. Determination of the Optimum Mass of the Solution for the Adsorption of Cu(II) Ions by Activated Carbon of Cassava Peel Waste

The analysis results concentration of Cu (II) in determining the optimum mass of the solution to the adsorption of metal ions Cu (II) activated carbon of cassava peel waste can be seen in Table 1. Based on Table 1, the relationship curve between adsorbent mass and %Cu (II) adsorbed on cassava peel activated carbon is obtained can be seen in Figure 1.

Table 1. Results concentration of Cu (II) in determining the optimum mass of the solution to the adsorption of metal ions Cu (II) activated carbon of cassava peel waste

| Adsorbent mass (mg) | Abs pH | Contact time (minutes) | C_i (mg/L) | C_eq (mg/L) | C_b (mg/L) | \( \frac{\chi}{m} \) (mg/g) | % Trapped |
|---------------------|-------|------------------------|------------|-------------|------------|----------------|-------------|
| 300                 | 0.0461| 5                      | 50         | 3.127       | 46.873     | 7.812          | 93.746     |
| 400                 | 0.0290| 5                      | 50         | 1.573       | 48.427     | 6.053          | 96.854     |
| 500                 | 0.0242| 5                      | 50         | 1.136       | 48.864     | 4.886          | 97.728     |
| 600                 | 0.0280| 5                      | 50         | 1.482       | 48.518     | 4.043          | 97.036     |

Figure 1. Relationship curve between adsorbent mass and % Cu(ii) adsorbed on cassava peel activated carbon

The effect of adsorbent amount is an important parameter because it can determine the capacity of the adsorbent during the addition of adsorbate concentration [15]. In Figure 1 it can be seen that the increase in adsorption that occurs at the adsorbent weight of 300 mg to 500 mg shows that the addition of an increasing number of adsorbents causes the surface of the adsorbent to be wider, so that more adsorbate is adsorbed [16]. The increasing weight of the adsorbent is proportional to the increase in the number of particles and the surface area of the adsorbent, causing the number of binding sites for metal ions to also increase and the absorption efficiency to increase [17].

The decrease in adsorption on the addition of 600 mg adsorbent mass was due to the metal Cu ions having been completely adsorbed. So that the addition of adsorbent mass does not affect the amount of metal ions adsorbed [18]. Under these conditions, the adsorption process was declared to have stopped because based on the value of %Cu that was adsorbed it had approached equilibrium where the number of adsorbate molecules bound to the adsorbent was getting smaller. The situation has reached the saturation point where the surface has been filled with adsorbate [19]. Based on the data in Table 1 and Figure 1, the optimum value for the variation of the mass of the solution using activated carbon of cassava peel as an adsorbent was at a mass of 500 mg which was able to adsorb 97.72% Cu(II) ions. The best adsorbent mass obtained was at a mass of 500 mg. This mass is different from the results of research by Adriansyah [19] who conducted a test of Cu metal adsorption using coffee rind as adsorbent.
3.2. Determination of the Optimum pH of the Solution for the Adsorption of Cu(II) Ions by Activated Carbon of Cassava Peel Waste

The analysis results concentration of Cu (II) in determining the optimum pH of the solution to the adsorption of metal ions Cu (II) activated carbon of cassava peel waste can be seen in Table 2.

The acidity (pH) is a factor that greatly affects the adsorption process of metal ions in solution, because the presence of H\(^+\) ions in solution will compete with cations to bind with active site. In addition, pH will also affect the ionic species present in the solution, so that it will affect the interaction between ions and the active site of the adsorbent [20]. Figure 2 Relationship between pH and % Cu(II) Adsorption on Cassava Peel Activated Carbon.

| Adsorbent mass (mg) | Abs (mg/L) | pH | Contacts time (Minutes) | C\(_i\) (mg/L) | C\(_eq\) (mg/L) | C\(_b\) (mg/L) | \(\frac{\chi}{m}\) (mg/g) | % Trapped |
|---------------------|------------|----|-------------------------|----------------|----------------|---------------|-------------------|-----------|
| 500                 | 0.0218     | 5  | 60                      | 50             | 1.073          | 48.927        | 4.892            | 97.854    |
| 500                 | 0.0183     | 6  | 60                      | 50             | 0.754          | 49.246        | 4.924            | 98.492    |
| 500                 | 0.0216     | 7  | 60                      | 50             | 1.054          | 48.946        | 4.894            | 97.892    |
| 500                 | 0.0617     | 9  | 60                      | 50             | 4.7            | 45.3          | 4.53             | 90.6      |

At pH 5 the adsorbent has an absorption capacity of 97.85%. The low absorption is due to the absorption of Cu (II) metal ions that do not work optimally in solutions that have an acidic pH value due to the concentration due to the H\(^+\) concentration being too high so that the negative functional group will react with H\(^+\) and prevent the binding of Cu (II) ions [21]. At pH 7 the absorption of the adsorbent decreased by 97.89%. The decrease in absorption at neutral pH because the ions can undergo a hydrolysis reaction in solution, it is not stable in the original ionic form and causes the adsorbent's ability to absorb to decrease [17].

Figure 2. Curve of the relationship between pH and % Cu(II) adsorbed on cassava peel activated carbon

The absorption of the adsorbent at pH 9 decreased significantly, namely 90.6% this is because at pH 9 or alkaline pH, Cu(II) ions will precipitates to form Cu(OH)\(_2\) which can prevent adsorption so that Cu(II) metal ions cannot be absorbed properly [22].

\[
\text{Cu}^{2+} (aq) + 2\text{OH}^-(l) \rightarrow \text{Cu(OH)}_2(s)
\]

Based on the above data, the optimum value for the variation of pH solution using cassava peel activated carbon adsorbent is at pH 6 which is able to adsorb as much as 98.49% Cu(II) ions.
The best adsorption pH obtained at pH 6 was different from the results obtained in the Cu metal adsorption test using corncob biomass adsorbent with the best Cu(II) metal adsorption conditions at pH 7 [10] and the optimum pH for Cu ion absorption with coffee husk biomass adsorbent was 4 [23]. Based on this, it can be said that the difference in optimum pH is due to the difference in the composition of the biomass used.

3.3. Determination of the Optimum Contact Time of the Solution for the Adsorption of Cu (II) Ions by Activated Carbon of Cassava Peel Waste

The analysis results concentration of Cu (II) in determining the optimum contact time of the solution to the adsorption of metal ions Cu (II) activated carbon of cassava peel waste can be seen in Table 3.

| Adsorbent mass (mg) | Abs pH | Contact time (Minutes) | C_i (mg/L) | C_eq (mg/L) | C_b (mg/L) | x_m (mg/g) | % Trapped |
|---------------------|--------|------------------------|------------|-------------|------------|------------|-----------|
| 500                 | 0.0131 | 6                      | 15         | 50          | 0.628      | 49,372     | 4.937     | 98.744    |
| 500                 | 0.0144 | 6                      | 30         | 50          | 0.779      | 49,221     | 4.922     | 98.442    |
| 500                 | 0.0127 | 6                      | 60         | 50          | 0.581      | 49,419     | 4.941     | 98.838    |
| 500                 | 0.0143 | 6                      | 90         | 50          | 0.767      | 49,232     | 4.923     | 98.466    |

Based on Table 3, the relationship curve between adsorbent contact time and %Cu (II) adsorbed on cassava peel activated carbon is obtained can bee seen in Figure 3.

![Figure 3. Curve of the Relationship between Contact Time and % Cu(II) Adsorbed on Cassava Peel Activated Carbon](image)

The results of the optimum contact time determination test show that the optimum contact time occurs at 60 minutes with an absorption capacity of 98.84%. Furthermore, in the 15th minute the absorption of Cu metal ions was 98.74% and in the 30th minute the absorption of Cu metal ions decreased by 98.44%. Before reaching the optimum contact time, the adsorption process in the 15th and 30th minutes there was an increase and decrease. This fluctuating value is due to the adsorption that occurs is reversible (back and forth) so that the absorption of the adsorbent becomes unstable on Cu(II) metal ions [24].

After reaching the optimum contact time the adsorption process experienced a decrease in absorption, namely at 90 minutes with an absorption capacity of 99.63%, this occurs because of the desorption or re-release of ions that bind to the adsorbent which is experiencing saturation where the pores in the adsorbent are fully filled [25].

The best adsorption time obtained was 60 minutes, which was different from the results obtained in the Cu metal adsorption test using a coffee rind adsorbent with the best condition for Cu(II) metal absorption being 100 minutes [23].
3.4. Determination of Adsorption Capacity with Variation of Cu(II) Metal Ion Concentration by Activated Carbon of Cassava Peel Waste

In this study, the Langmuir adsorption isotherm equation was used to apply to the adsorption of copper ions by activated charcoal of cassava peel. The Langmuir linearity for Cu(II) absorption at various concentrations is shown in Figure 4.

**Figure 4.** Langmuir Linearity Curve for Cu(II) Absorption at variations in concentration

Langmuir isotherm is used with the assumption that the layer formed is a monolayer layer where the adsorbent bond with the adsorbate is strong enough due to the formation of a chemical bond [26]. Based on the linearity curves for uptake of Cu (II) concentration variation correlation coefficient $R^2 = 0.9917$ with a maximum adsorption capacity $(\alpha) = -51.813 \text{ mg/g}$. In addition, the value of the Langmuir constant $(\beta)$ is -0.34.

**Figure 5.** Freundlich Linearity Curve for Cu(II) Absorption at Variations in Concentration

The Freundlich isotherm is used with the assumption that the multilayer layer in which the bond between the adsorbent and the adsorbate occurs due to Van der Waals forces so that the bond is not too strong [26]. Based on the linearity curves for uptake of Cu (II) concentration variation correlation coefficient $R^2 = 0.9996$ with a maximum adsorption capacity $(k) = 26.792 \text{ L/g}$, and n value of 6,301 which is a constant Freundlich [27]. The value of n indicates the adsorption characteristics. The adsorption process occurs very well if the n value is 2-10, sufficient if the value is 1-2, and bad if the value is <1 [29]. In this study, the value of n was 6.301 and indicated that the adsorption process was very good.

Adsorption isotherm pattern for each adsorbent mass can be determined by looking at the value of the coefficient of determination ($R^2$) on the adsorption equation. The largest correlation coefficient value indicates that the adsorption process using the adsorbate is in accordance with the assumed characteristics. From the two graphs above, it can be seen that the adsorption of Cu(II) metal ions by activated carbon of cassava peel waste is more likely to follow the Freundlich isotherm equation than Langmuir, it can be seen from the value of the coefficient of determination $R^2$, which is 0.9996 (close to number 1). This value assumes that the adsorption that occurs is physical adsorption that forms a
multilayer layer and the activated carbon used has a very large surface area so that all Cu(II) metal ion molecules can be adsorbed by forming two or more layers [28].

Conclusion
The conclusions of this study are: that the best conditions (optimal conditions) were obtained with the addition of 0.5 grams of active carb from cassava peels. The percentage of Cu ion adsorption in these conditions was 97.72%, at pH 6 and a contact time of 60 minutes and the absorption capacity was 98.49%. The maximum adsorption capacity of cassava peel activated carbon to Cu(II) ions at the optimum condition was determined based on the Langmuir and Freundlich isotherm equations. The results obtained were -51.813 mg/g and 26.792 mg/g, respectively.

Acknowledgement
The author would like to thank the Teacher Training Education Faculty of Tadulako University.

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