



KINETICS OF TOFU PULP AS COPPER METAL ADSORBENT ON THE WASTE OF THE SILVER CRAFT INDUSTRY

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Abstract

Liquid waste originating from the silver crafts industry is heavy metal waste that is dangerous for the surrounding environment, one of which is copper (Cu). Copper is a heavy metal that can have toxic effects on human health. Based on these problems, processing is needed to reduce the pollutants produced. One of the technologies used is adsorption using a natural adsorbent that is efficient and easy to obtain, namely tofu dregs. Adsorption using tofu dregs is because tofu contains protein. This protein has the ability to absorb amino acids which form two charged ions (zwitter ions). Toxic metals can be bound to proteins as metallothionein. This research aims to determine the efficiency of using tofu dregs as an adsorbent for copper metal (Cu) and to determine the effect of contact time and adsorption isotherms on copper metal adsorption. 1 gram of tofu dregs was used with 25 ml of waste and then contacted with a magnetic plate stirrer at varying times of 30, 60, 90, 120 and 150 minutes. The concentrations of all copper metals were analyzed using an Atomic Absorption Spectrophotometer. Next, each copper metal content was calculated using a copper metal standard calibration curve. Identification using an Infrared Spectrophotometer showed that the tofu dregs waste in this study contained the functional groups -OH, N-H (stretching), C-H (aliphatic), C=O, -OH (bending vibration), and C-O. From the research it was found that the copper metal content in the sample was 19.5979 ppm. The optimum contact time for copper metal adsorption is 120 minutes, which results in an optimum adsorption efficiency for copper metal, namely 54.88%. The isotherm equation that corresponds to the adsorption of copper metal using tofu dregs waste is the Freundlich isotherm with a Kf value of 0.072 L/g and a 1/n value of 2.366. This shows that the adsorption of copper metal occurs in a multilayer layer on the surface of the tofu dregs adsorbent, which illustrates the occurrence of physical adsorption. Tofu dregs waste is an effective adsorbent for reducing copper metal levels in silver craft waste.

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INTRODUCTION

Ungga Village, Praya Barat Daya District, Central Lombok Regency is one of the silver handicraft industry villages that contributes to generating the largest Local Original Income (PAD) in Central Lombok Regency. Data from Kompas (2017), currently there are around 200 silversmiths in Ungga Village. As handicraft commodities increase, so does the waste produced. Liquid waste originating from silver crafts is heavy metal waste that is harmful to the surrounding environment, one of which is copper metal (Cu) contained in its wastewater. Cu waste appears in dyeing using hydrochloric acid (HCl) which is acidic and serves to dissolve impurities attached to silver after the forging process (Andaka, 2008). The toxicity possessed by copper metal will work and show its effect when this metal has entered the organism's body in large quantities or exceeds the tolerance value of the organism. In humans in high doses can cause symptoms of kidney, liver,

spurtber, dizziness, weakness, anemia, coma, and can cause sufferers to die (Palar, 2012). Based on the Regulation of the Minister of Environment of the Republic of Indonesia Number 5 of 2014 concerning Wastewater Quality Standards, it is determined that the threshold of copper metal in wastewater should not exceed 0.5 mg / L. If copper metal waste with high concentrations is disposed of without treatment, then this waste will seep into the ground and pollute water sources in residential areas because heavy metal waste is difficult to degrade.

Copper metal (Cu) is the best conductor of electricity after silver, so it is widely used in the field of electronics. Copper can also form *alloys* with various other metals, for example: zinc, tin, or lead (Cu-Zn-Sn-Pb) in brass form which is often used in household appliances. Copper compounds are widely used in the paint industry as antifoling, insecticide and fungicide industries, as catalysts, batteries, electrodes, sulfur attractors, and as pigments and moss growth inhibitors (Apriliani, 2010). Under normal conditions, copper in waters is found in the form of CuCO_3 and Cu(OH)_2 compounds. If in the waters there is an increase in copper solubility beyond the proper limit, there will be a biomagnification event on aquatic biota. Based on this, it is important to treat wastewater to remove or reduce copper metal levels in the environment. Handling copper metal pollutants has been carried out by various methods, including: precipitation, ion exchange, electrodialysis, reverse osmosis, ultrafiltration, and adsorption (Handoko, et al., 2013).

The adsorption method using adsorbents of natural materials often called biosorption is attracting attention at the moment. This method is considered more effective, economical and often used in wastewater handling (Selvi, et al., 2011). The adsorption process is an effective purification and separation technique used in industry because it is considered more economical in wastewater treatment and is a technique that is often used to reduce metal ions in water. Various adsorbents can be used for liquid waste treatment, including activated carbon, alumina, silica gel, zeolite and other materials that have activity to absorb a chemical such as tofu dregs. Tofu pulp is only used by farmers as animal feed and raw material in making tempeh gembus. There is another potential for the use of tofu dregs, namely as an adsorbent.

Tofu industry liquid waste contains a large amount of carbohydrates, fats and proteins. Organic molecules contained in industrial liquid waste know broadly undergo an overhaul, especially carbohydrates, fats and proteins contained therein carried out by decomposing microorganisms. Complex organic matter in the form of carbohydrates, fats and proteins is first converted into simpler forms of compounding namely glucose, glycerol, fatty acids and amino acids. Amino acids that are the result of protein remodeling will be oxidized to nitrogen, ammonia (NH_3) and carboxyl compounds. The compound (NH_3) will be oxidized again to nitrite (NO_2). When oxygen is available, it is oxidized again to nitrate (NO_3) (Pelczar and Chan, 1996).

The content of nitrogen and other nutrients at high concentrations in water will accelerate the growth of aquatic plants. Such conditions will gradually cause the death of biota in water (Bahri, 2006). Widayat, W., Suprihatin, Herlambang, A, 2010, mentioned that non-ionized ammonia is very toxic to fish. Fish death can occur in the presence of 0.1 mg/L to 10,000 mg/L of ammonia in waters. Nitrates are more toxic than nitrites, can cause kidney damage and cancer. Nitrates can also bind to hemoglobin and cause oxygen deprivation in infants also called methemoglobinemia. However, nitrites can react with amines chemically or enzymatically to form nitrosamines with very strong carcinogen properties (Wanielista and Chang, 2008).

The attractive force on a solid is divided into two types, namely physical force and chemical force, which respectively produce physical adsorption (*physisorption*) and chemical adsorption (chemisorption) (Roto; Dahlia; and Agus; 2015). Physical adsorption occurs due to the Van der Waals force. In physical adsorption, the attractive force between fluid molecules and molecules on the surface of solids is smaller than the attractive force between fluid molecules so that the attractive force between adsorbate and the adsorbent surface is relatively weak. Chemical adsorption occurs due to the chemical bond formed between the adsorbate molecule and the adsorbent surface. Chemical bonds can be covalent bonds or ionic bonds. The bonds formed were so strong that the original species could not be determined. The strong chemical bond formed causes the adsorbate not easily desorbed. Chemical adsorption begins with physical adsorption where the adsorbate is close to the surface of the adsorbent through Van der Waals forces or hydrogen bonds then attached to the surface by forming chemical bonds which are usually covalent bonds (Shofa, 2012).

This research is intended to develop the potential of processing waste originating from the tofu processing industry so as not to harm the environment. Tofu waste can be used to bind ions or metals present in water because waste comes from tofu industry waste which still has the same properties as finished tofu even though it has been destroyed. The use of tofu waste as an absorber (adsorption) because tofu contains proteins that have the absorption of amino acids that form zwitter ions (two-charged). Proteins that have active sides (clusters) can bind metal ions or other compounds. Harmful metals such as cadmium, lead, mercury, chrome, and arsenic that are toxic can be bound with proteins as metalotionein (Nohong, 2012).

Based on previous research conducted by Nohong (2012), tofu waste powder can reduce chromium (Cr) metal levels in landfill leachate water by 98% and reduce iron metal (Fe) levels in landfill leachate water by 95%. Tripathi (2015) uses tofu pulp as an adsorbent in reducing lead heavy metal (Pb) which shows the results that every 1 gram of tofu pulp can reduce Pb ions by 29.85 mg. Research by Taufieq (2011) showed the results that tofu pulp can reduce the nickel (Ni) metal content in red and yellow padzolic soil in Soroako from 2.6% to 1.45%.

The use of tofu pulp into natural adsorbents has good prospects and is economical to develop. Therefore, this tofu pulp adsorbent is expected to be an added value and increase its carrying capacity to the environment. By using the *batch* method and metal absorption analysis using an Atomic Absorption Spectrophotometer, it is hoped that this research can be used as a contribution to science in waste management efforts so as to reduce environmental pollution caused by the presence of harmful metal ions.

METHOD

1. Analysis of Copper Metal Levels in Silver Handicraft Industry Waste.

A total of 25 mL of liquid waste samples of the silver handicraft industry in Ungga Village were put into bottles and labeled. The initial content of copper metal in the sample was analyzed using an Atomic Absorption Spectrophotometer.

2. Preparation of Tofu Dregs Waste Adsorbent.

In adsorbent preparation, tofu pulp is deformed into tofu waste powder. The steps for making tofu waste powder are as follows:

- a. Solid tofu waste dried at room temperature
- b. Dry tofu waste is heated in an oven at 60°C for 14 hours

- c. Dried tofu waste is mashed with a blender
- d. Tofu waste powder is sifted using a 60 mesh sieve
- e. Tofu waste powder is ready for use
- f. The results of tofu waste powder that have become adsorbents are stored in aluminum foil.

3. Tofu Dregs Adsorbent Characterization

a. Moisture Content Determination

A total of 2.0 grams of adsorbent was heated at 110°C for 2 hours, cooled and weighed until a constant weight was obtained.

b. Determination of Functional Groups

The determination of functional groups contained in tofu pulp adsorbents is carried out using an Infrared Spectrophotometer

4. Adsorption Experiment

Tofu waste powder was weighed each with a mass of 1 gram and put into 5 pieces of erlenmeyer each. Then 25 mL of silver craft industry wastewater samples were inserted into each erlenmeyer. Erlenmeyer was placed on a magnetic plate stirrer at room temperature during adsorption contact time variations of 30, 60, 90, 120 and 150 minutes. After that the mixture is separated by filtering using filter paper. The filtered filtrate is taken 10 mL and added 1 drop of nitric acid as a preservative so that there are no changes in the composition of the solution and then the copper metal content is measured with an Atomic Absorption Spectrophotometer.

5. Determination of Adsorption Isotherms Equation

The isotherm equation is the basis for determining the adsorption system, because it describes the interaction that occurs between the adsorbate and the adsorbent. Determination of adsorption isotherm equation in this study using adsorbate concentration variation data. In this study, there were two types of isotherms tested, namely Langmuir isotherms and Freundlich isotherms.

Langmuir isotherms show that adsorbents have a homogeneous surface by a monolayer adsorption process. Langmuir's Isotherms Equation is (Tchobanoglous et al, 2014):

$$\frac{C_e}{q_e} = \frac{C_e}{q_{max}} + \frac{1}{K_L q_{max}} \dots \dots \dots (3)$$

Where: C_e is the final concentration of adsorbate at equilibrium after adsorption (mg/L), q_e is the mass of the adsorbed substance (adsorbate) per adsorbent mass (mg/g). q_{max} is the maximum adsorption capacity of the adsorbent (mg/g) and K_L is the Langmuir adsorption constanta (L/mg). The Langmuir isothermic constant can be determined by plotting C_e/q_e data on the y-axis with respect to C_e .

Freundlich isotherms show that adsorbents have a heterogeneous surface and each molecule has different absorption potential for compounds by a multilayer adsorption process. Freundlich adsorption isotherm equation:

$$q_e = K_F C_e^{1/n} \dots\dots\dots(4)$$

Where: q_e is the mass of the adsorbed substance (adsorbate) per adsorbent mass (mg/g); K_f is the relative adsorption capacity of Freundlich; C_e is the final concentration of adsorbate at equilibrium after adsorption (mg/L) and $1/n$ is the intensity of the Freundlich parameter.

The constants on Freundlich isotherms (K_f and n) can be determined by plotting data on a graph, where the q_e log on the y-axis is against the C_e log on the x-axis, and the equation (5) is obtained (Tchobanoglous et al., 2014):

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \dots\dots\dots(5)$$

A linear curve is formed between C_e and C_e/q_e for Langmuir's equation and between $\log C_e$ and $\log q_e$ for Freundlich's equation. The coefficient of determination (R^2) obtained from each equation is used to assess the isothermic equation corresponding to this experiment.

RESULTS AND DISCUSSION

1. Copper Metal Content in Silver Craft Waste

The results of testing silver craft wastewater samples in Ungga Village, Central Lombok using Atomic Absorption Spectrophotometry (AAS) obtained copper metal concentrations with an average of 19.5979 ppm. These results are obtained from calculations on the standard curve of copper solution which has the equation $Y = 0.10937X + 0.00075238$ with a regression coefficient $r^2 = 0.9999$. The copper concentration in the wastewater is then used as the initial concentration of copper before treatment.

2. Preparation of Tofu Dregs Waste Adsorbent.

The result of the preparation of the tofu dregs waste adsorbent obtained is 250 grams of dry tofu dregs waste from 1 kilogram of tofu dregs waste. The dried tofu pulp obtained is then mashed to obtain a uniform size and stored in a closed container to avoid contact with air.

3. Tofu Dregs Adsorbent Characterization

a. Moisture Content Determination

In general, the moisture content of a material to be used as an adsorbent material is a maximum of 10%. If the moisture content of tofu pulp is increasing, the ability of the adsorbent to absorb the adsorbate will be smaller because the water molecules cover the pores of the adsorbent. Determination of moisture content is done gravimetrically by dry the ingredients in the oven at 110°C until a constant weight is obtained. In the study, 2.0 grams of tofu pulp adsorbent were taken and then dried in the oven using a temperature of 110°C and obtained a moisture content of 5.03%. These results show that tofu pulp has the ability as an adsorbent.

b. Determination of functional groups

Determination of functional groups contained in adsorbents is carried out using a FTIR Spectrophotometer. The FTIR spectrum of tofu pulp adsorbents shows several

absorption patterns, namely absorption that appears in the 3336.85 cm^{-1} area which is the absorption of the hydroxyl group (-OH), the $3282.84 - 3296.35\text{ cm}^{-1}$ area there is amine absorption which is in a position close to each other with -OH absorption. Because amines have weaker hydrogen bonds and some N-H is less polar, uptake is less intensive than with -OH. Absorption from C-H (*stretching*) is in the area of $2858.51 - 2929.87\text{ cm}^{-1}$. Absorption in the area of 1635.64 cm^{-1} shows that the vibration of C=O peptide is carbonyl absorption. Absorption in the area of 1371.39 cm^{-1} is the absorption area of bending vibration of carboxylic acid, while in the absorption area of 1047.35 cm^{-1} shows C-O vibration of carboxylic acid.

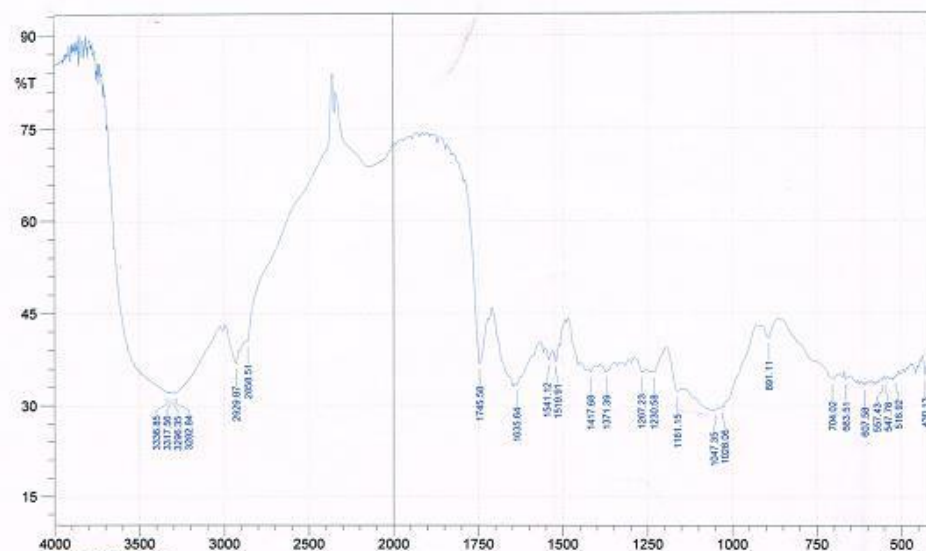


Figure 1. Tofu Dregs Waste FTIR Results

4. Adsorption Process

The determination of the adsorption equilibrium time aims to determine the minimum time required by the adsorbent in absorbing the adsorbate to the maximum until the equilibrium state is reached. The adsorption process takes place continuously as long as there is no equilibrium. Therefore, to find the equilibrium distribution between the adsorbent and the adsorbate, it is necessary to vary the contact time.

A total of 25 mL of wastewater samples were then contacted with 1 gram of tofu pulp adsorbent at a variation of contact time of 30, 60, 90, 120, and 150 minutes. Adsorbent contact on silverware wastewater samples containing copper is carried out with a *batch* system, which is mixing adsorbents in a fixed amount of solution and observed changes in quality at certain intervals. This study was conducted on samples with a copper concentration of 19.5979 ppm.

The decrease in copper concentration in the waste samples ranged from 19.41% to 54.88%. According to Manocha (2013), adsorption is a phenomenon that is closely related to the surface where interactions between liquid or gas molecules are involved with solid molecules. This interaction occurs due to the attraction of atoms or molecules that cover the surface. The adsorption capacity of tofu pulp waste depends on the pore type and the number of surfaces that may be used for adsorption. The concentration of copper in contact time variation is shown in Figure 2.

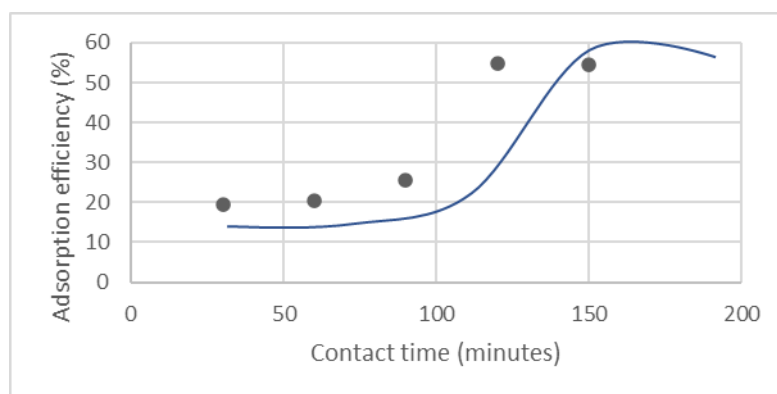


Figure 2. The effect of contact time on copper adsorption efficiency

From Figure 2 it can be seen that the best copper adsorption efficiency in silver craft waste samples is at a contact time of 120 minutes which is 54.88%. Sufficient contact time is required by tofu pulp waste adsorbents in order to adsorb copper optimally. The longer the contact time, the more chances the particles have to come into contact with the copper metal and bond in the pores of the adsorbent. But at a certain contact time, the adsorption efficiency decreases.

Decreased adsorption efficiency is possible due to the desorption process or re-release of adsorbate during stirring. Desorption occurs due to the saturated adsorbent surface. In the saturated state, the adsorption rate is reduced so that the contact time no longer has an effect. Metal adsorption increases at long reaction times because the number of active sites available on the surface is unsaturated (Roto; Dahlia; and Agus; 2015). In copper metal adsorption, after 120 minutes the amount of absorbed metal did not undergo significant changes.

5. Determination of Adsorption Isotherms Equation

The adsorption isotherm equation is very important in adsorption systems. In general, adsorption isotherms describe how adsorbates interact with adsorbents (Khodaie et al., 2013). To study the phosphate adsorption isotherm model on corn husks, the isotherms equation tested is the Langmuir and Freundlich isotherm equations. The data used in the determination of this adsorption isotherm are data from adsorbate concentration variation experiments (Tchobanoglous et al., 2014). Figure 3 and Figure 4 show Langmuir and Freundlich isotherm plots for copper metal adsorption using tofu pulp adsorbents.

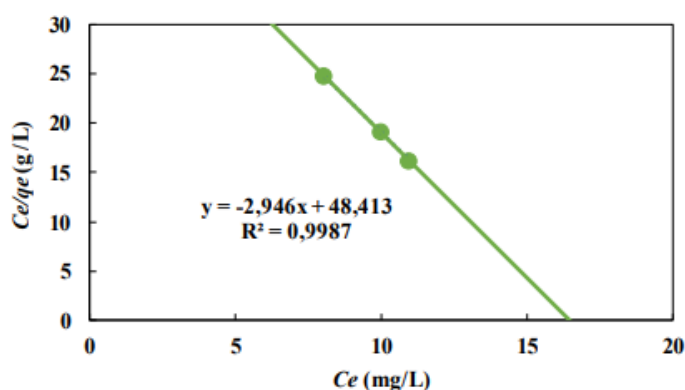


Figure 3. Langmuir isotherms for copper metal adsorption using tofu pulp

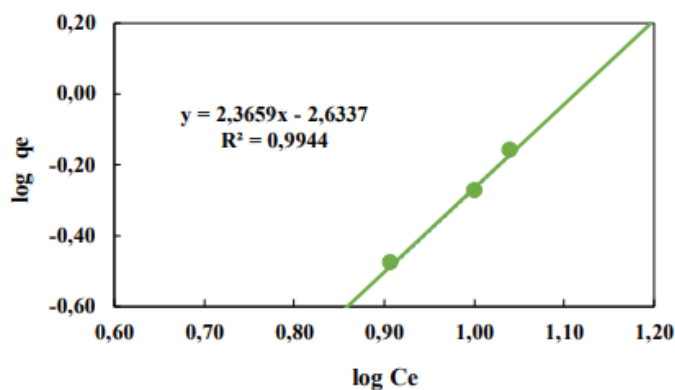


Figure 4. Freundlich isotherms for adsorption of copper metal using tofu pulp

Table 1. Copper metal adsorption isotherm constant using tofu pulp

Isotherms Equation	Constant	Value
Langmuir Isotherms	R^2	0,9987
	K_L	-0,061
	q_m	-0,34
Freundlich Isotherms	R^2	0,9944
	K_f	0,072
	$1/n$	2,366

From Table 1 it can be seen that based on the comparison of the value of the coefficient of determination (R^2) between Langmuir isotherms and Freundlich isotherms, it is found that the R^2 value of Langmuir isotherms is greater and closer to 1 than Freundlich isotherms. However, Langmuir isotherms obtained maximum capacity values (q_m) and Langmuir constant (K_L) values negative values indicating that Langmuir isotherm models are not sufficient to describe the interactions that occur in copper metal adsorption using tofu pulp adsorbents. This means that Freundlich isotherms are more suitable following the equilibrium model for adsorption of metal phosphate using tofu pulp adsorbents.

Based on this it can be assumed that the active side of the tofu pulp surface in adsorbing copper metal is heterogeneous and adsorption can occur in a timely manner. multilayer. In addition, the selection of Freundlich isotherms in this study shows that the adsorption mechanism that occurs is physical adsorption where it can be assumed that the interaction occurs by phosphate entering the adsorbent pores through electrostatic interaction or interacting with weak bond energy, namely Van der Waals bonds (Atkins et al., 2018).

According to Atkins et al. (2018), the value of n in Freundlich isotherms that are in the range of 1-10 in the adsorption process is said to be beneficial. In addition, the value of $1/n$, which indicates the intensity of the adsorption, smaller than 1 also indicates favorable adsorbent conditions (favourable) to be used as an adsorbent because the interaction between the adsorbate and the adsorbent is strong so that the adsorbate is not easily separated from the adsorbent surface. The n value in this study is 0.423 with a value of $1/n$ of 2.366 so that tofu pulp as an adsorbent in the copper metal adsorption process is less

profitable (unfavourable). This illustrates the weak interaction between the adsorbate (copper) and the tofu pulp adsorbent so that the adsorbate easily escapes from the adsorbent surface and causes the adsorption process to be less than optimal.

CONCLUSION

The utilization of tofu pulp waste as a copper metal adsorbent in the silver craft industry has been carried out. From the study, it was found that the copper metal content in the sample was 19.5979 ppm. The optimum contact time in copper metal adsorption is at a contact time of 120 minutes which results in an optimum adsorption efficiency in copper metal of 54.88%. Identification using an FTIR spectrophotometer shows that in tofu dregs waste there are functional groups -OH, N-H (*stretching*), C-H (aliphatic), C=O, -OH (bending vibration), and C-O. The isothermic equation corresponding to copper metal adsorption using tofu pulp waste is Freundlich isotherm with a Kf value of 0.072 L/g and a value of 1/n 2.366. This shows that copper metal adsorption occurs in the multilayer layer of the tofu pulp adsorbent surface which describes the occurrence of physical adsorption. Tofu pulp waste is an effective adsorbent to reduce copper metal levels in silver craft waste.

RECOMMENDATIONS

Further research can be done for adsorption of other heavy metals as well as using tofu pulp waste that has been modified with chelating material that more effective in adsorbing metals.

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