Utilization Of Banana Peel And Water Hyacinth Leaves As Adsorbent For Removal Of Copper From Wastewater

Riana Ayu Kusumadewi, Asih Wijayanti, Rositayanti Hadisoebroto*

Abstract: This research aims to compare the performance of adsorption of copper heavy metal by banana peel and water hyacinth leaves as adsorbent. Heavy metal pollution is one of the most pervasive environmental problems globally. Adsorption of heavy metals on conventional adsorbents such as activated carbon, that produced by carbonizing organic materials, have been used widely in many applications. Some of the low cost adsorbents that were tested in the study for the dye sorption process were banana peel and water hyacinth leaves, focus on the effect of mixing speed, contact time, and weight of the adsorbent on the efficiency of Cu removal. A maximum removal of 99.55% was observed at the adsorbent mass of 5 grams and contact time of 30 minutes using banana peel adsorbent, while using water hyacinth leaves, a maximum removal of 98.39% was observed at mass of the adsorbent of 15 grams and contact time of 60 minutes. Both the observed adsorbents fit the Langmuir isotherm model of adsorption. The quality of water produced after the adsorption process using the adsorbent of banana peel and water hyacinth leaves has met the quality standard according to the Regulation of the Minister of Environment of the Republic Indonesia Number 5 of 2014 concerning quality standards for wastewater in the paint industry. It could be concluded that the organic waste like banana peel and water hyacinth leaves were promising biosorbents and solving the heavy metal pollution in environment.

Index Terms: Adsorption, banana peel, contact time, copper removal , mixing speed, water hyacinth leaves, weight of adsorbent

1 INTRODUCTION

The dye and heavy metal are two principal concomitant pollutants in industrial wastewaters, posing a serious threat to public health and the environment [1]. Heavy metal pollution is one of the most pervasive environmental problems globally [2]. Heavy metals are produced in large amounts during industrial activities and contaminate the environment. Metal ions are non-biodegradable and many of them are soluble in aqueous media and easily available for living organisms. Heavy metals account for a number of disorders in plants and animals and their removal from aqueous media is an important and challenging task [3]. Heavy metals pollution occurs in much industrial wastewater such as that produced by metal plating facilities, mining operations, battery manufacturing processes, the production of paints and pigments, and the ceramic and glass industries. This wastewater commonly includes Cd, Pb, Cu, Zn, Ni and Cr. Whenever toxic heavy metals are exposed to the natural eco-system, accumulation of metal ions in human bodies will occur through either direct intake or food chains [4]. Dyes are potential environmental contaminants, liberated in the aqueous streams by the wastewaters of different industries like textiles, pulp, leather, cosmetics, food and paper industries [5]. Adsorption has been proved to be excellent approach for heavy metals and semimetals removal from wastewaters and polluted soils [6].

Numerous methods are available for the removal of heavy metals from aqueous solutions including chemical precipitation, ion exchange, ultra-filtration, reverse osmosis, and adsorption. However, these methods have some limitations due to the production of secondary wastes, large quantity of sludge formation and high operational costs. In contrast adsorption is more advantageous than the other methods due to its simple operation design with sludge free environment and low cost [3]. Heavy metals should be prevented from reaching the natural environment. In order to remove toxic heavy metals from water systems, conventional methods have been used such as chemical precipitation, coagulation, ion exchange, solvent extraction and filtration, evaporation and membrane methods. Adsorption of heavy metals on conventional adsorbents such as activated carbon have been widely used in many applications as an effective adsorbent, and the activated carbon produced by carbonizing organic materials is the most widely used adsorbent. However, the high cost of the activation process limits its use in wastewater treatment applications [4]. Treatment processes for heavy metal removal from wastewater include precipitation, membrane filtration, ion exchange, adsorption, and co-precipitation. Studies on the treatment of effluent bearing heavy metals have revealed adsorption to be a highly effective technique for the removal of heavy metals from waste stream and activated carbon has been widely used as an adsorbent. Despite its extensive use in water and wastewater treatment industries, activated carbon remains an expensive material. Adsorption processes are being widely used by various researchers for the removal of heavy metals from waste streams and activated carbon has been frequently used as an adsorbent. Despite extensive use in water and wastewater treatment industries, activated carbon remains an expensive material. In recent years, the need for safe and economical methods for the elimination of heavy metals from contaminated waters has necessitated research interest toward the production of low cost alternatives to commercially available activated carbon. Therefore, there is an urgent need that all possible sources of agro-based inexpensive adsorbents should be studied in detail [7]. Nowadays, it is a

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growing demand to find out a locally available, low cost, environmentally friendly and efficient adsorbent for the removal of heavy metal from industrial and municipal waste [8]. Biosorbers have recently attracted significant attention for wastewater treatment [9]. Banana is one of the world’s most important crops grown by more than 130 countries. India, China, Uganda, Philippines, Ecuador, Brazil, Indonesia, Columbia, Cameroon and Ghana were the top ten bananas producing countries in the world in 2012. Several research groups have used raw and chemically treated banana peels and banana stalks for the removal of toxic heavy metal ions from aqueous solutions and industrial wastewater [3]. The huge quantity of banana peels waste from juice industries and fruit markets every day is indicating a potential bio-resource which is major task for environment safety. It can be converted into useful material instead of being dumped in landfill site, and reduce the environment and economic problems [10]. Study concluded that banana peels, a waste material, have good potential as an adsorbent to remove toxic metals like lead and cadmium from water [11]. Some of the low cost adsorbents that are tested for the dye sorption process are banana peel and water hyacinth. Water hyacinth originated in the American tropics and spread to all tropical climate countries. In India, they can be found in large water areas in the Kerala backwaters, ponds and lakes. Due to vegetative reproduction and vigorous growth rate of this plants, it dramatically impacts water flow, blocks sunlight from reaching native aquatic plants, and starves the water of oxygen, often killing fish and also acts as a prime habitat for mosquitoes [12]. In the recent past, some researchers emphasize using aqua vascular weed for removal of heavy metals like water hyacinth. Water hyacinth is grown abundantly throughout the tropical and subtropical region in the world and also available in Indonesia [8]. Water hyacinth (Eichornia crassipes) is an aquatic plant that can grow very rapidly and free-floating over the water surface. The invasive potential of this plant has created massive environmental problems in many areas. It has exhibited extremely high growth rates and the coverage of an entire aquatic body by forming thick compact carpet. This thick carpet blocks the passage of sunlight underwater so that the solubility of oxygen in water decrease, several problems including destruction of eco-systems, irrigation problems and as a mosquito breeding place leading to increase in mosquito population [13].

2 RESEARCH METHODOLOGY

2.1 Location and Time of Study
Screen printing wastewater samples were taken from home industries in Krendang village, Tambora Sub-district, West Jakarta. This research was conducted in Environmental Engineering Laboratory, Universitas Trisakti, from February to July 2019.

2.2 Working Method
At the beginning of the research, analysis of heavy metals contained in screen printing wastewater was conducted. These heavy metals include copper (Cu), zinc (Zn), titanium (Ti), total chrome (Cr), cadmium (Cd), mercury (Hg), and lead (Pb). Heavy metals concentration analysis was tested on several colors of screen printing wastewater, i.e. black, white, red, yellow, and blue. From the results of this analysis, obtained that blue wastewater contains the highest heavy metals contents compared to other heavy metals content, with Cu concentration of 3.532 mg/L. The concentration of Cu in wastewater does not meet the quality standard according to the Regulation of the Minister of Environment of the Republic of Indonesia Number 5 of 2014 concerning quality standards for wastewater in the paint industry. The permissible Cu concentration is less than 0.8 mg/L. Activation of The Adsorbent using Banana Peel and Hyacinth LeavesBanana peel and hyacinth leaves were washed by distilled water and running water. After banana peel and hyacinth leaves were clean, the next process was cutting banana peel and water hyacinth leaves into small pieces and dried in the sun for 5 days to reduce the moisture content attached to them. After drying, banana peel and hyacinth leaves were heated at 400°C for 2 hours until charcoal was formed. After that, banana peel and hyacinth leaves were smoothed into a uniform size and sieved with a 100 mesh sieve. Then those refined charcoal were put into the desiccator. The banana peel and hyacinth leaves which have been carbonized were then activated using a solution of H₂SO₄ with concentration of 20%. The ratio of adsorbates and adsorbents were 2 mL : 1 gram. Activation of the adsorbents were done by soaking the banana peel and hyacinth leaves into a H₂SO₄ solution for 24 hours. Then the activated carbon was washed with distilled water and dried in the oven at 110°C for 2 hours. This research aims to compare the performance of adsorption of heavy metal Cu by banana peel and water hyacinth leaves as adsorbent. The effect of mixing speed, contact time, and weight of the adsorbent on the efficiency of Cu removal with adsorbent of banana peel and water hyacinth leaves would be analyzed. At the beginning of the experiment, the optimum mixing speed and contact time in removing Cu were determined. The variations in mixing speed were 100, 150, and 200 rpm with contact times of each mixing of 30, 60, and 90 minutes, with a mass of 10 grams of adsorbent. Furthermore, determination of the optimum adsorbent mass and contact time in removing Cu were conducted. The mass variations of the adsorbent used were 10, 15, and 20 grams with contact times of each mass of the adsorbent of 30, 60, and 90 minutes.

The percentage of adsorption of Cu was calculated according to the following equation [3]:

\[
\text{%Removal} = \frac{C_0 - C_e}{C_0} \times 100\% \tag{1}
\]

Where \(C_0\) is the initial Cu ion concentration (mg/L) and \(C_e\) is the final Cu ion concentration (mg/L).

Derived empirically in 1912, the Freundlich isotherm is defined as follows [14]:

\[
\frac{x}{m} = K_f C_e^{1/n} \tag{2}
\]

Where \(x/m\) is mass of adsorbate adsorbed per unit mass of adsorbent (mg adsorbate/g activated carbon), \(K_f\) is Freundlich capacity factor (mg/g)(L/mg)\(1/n\), \(C_e\) is equilibrium concentration of adsorbate in solution after adsorption (mg/L), and \(1/n\) is Freundlich intensity parameter.

The constants in the Freundlich isotherm can be determined by plotting log \((x/m)\) versus log \(C_e\) and making use of the linear form of Eq. 2 rewritten as [14]:

\[
\log\left(\frac{x}{m}\right) = \log K_f + \frac{1}{n} \log C_e \tag{3}
\]
Derived from rational considerations, the Langmuir adsorption isotherm is defined as [14]:

\[ \frac{x}{m} = \frac{K_L b C_e}{1 + b C_e} \]  

(4)

Where \( x/m \) is mass of adsorbate adsorbed per unit mass of adsorbent (mg adsorbate/g activated carbon), \( K_L \) and \( b \) are empirical constants, and \( C_e \) is equilibrium concentration of adsorbate in solution after adsorption (mg/L). The constants in the Langmuir isotherm can be determined by plotting \( C_e/(x/m) \) versus \( C_e \) and making use of the linear form of Eq. 4 rewritten as [14]:

\[ \frac{C_e}{(x/m)} = \frac{1}{K_L b} + \frac{1}{K_L C_0} \]  

5 Langmuir equation can also be used to calculate a dimensionless equilibrium parameter, \( R_L \) also known as the separation factor and can be expressed as [15]:

\[ R_L = \frac{1}{1 + b C_0} \]  

(6)

Where \( C_0 \) is the initial concentration of copper (mg/L).

3 RESULTS AND DISCUSSION

3.1 Effect of Mixing Speed and Contact Time on Cu Removal Efficiency with Banana Peel and Water Hyacinth Leaves as Adsorbents

Contact time is one of the effective factors in batch adsorption process [15]. In this stage, all of the parameters except mixing speed and contact time, including mass of adsorbent (10 grams), initial copper concentration (3.532 mg/L), were kept constant. The effect of mixing speed and contact time on copper adsorption efficiency showed in Fig. 1.

As it is shown in Fig. 1., a maximum Cu removal efficiency, i.e. 98.19%, was observed at mixing speed of 100 rpm and contact time of 60 minutes using banana peel adsorbent, while using water hyacinth leaves adsorbent, a maximum Cu removal efficiency, i.e. 97.4%, was observed at mixing speed of 200 rpm and contact time of 90 minutes. The increase in heavy metal removal with increase in time is due to the higher interaction between the sorbent surface and metal ions [3]. The increased uptake of heavy metal ions with the passage of time might be due to the increased access of ions to active sites on the surface of adsorbent, resulting in an increase in the adsorption rate [10]. For banana peel adsorbent, at 90 minutes contact time, the efficiency of Cu removal decreased after increasing at 60 minutes contact time, so the adsorption phase reached to equilibrium at 60 minutes contact time. The contact time is one of the factors that influence the adsorption process, because through the contact process, adsorption occurs as a result of physical interaction (adhesion) between the adsorbate and the adsorbent [16]. According to research conducted by Herawati and Ngibad [17], the optimum contact time of textile waste dyes varied between 45 minutes and 75 minutes. This could be due to many facets adsorbent initially empty, so that the tendency of the solution to be adsorbed into the higher adsorbent. With increasing contact time, the amount of adsorbate adsorbed on the surface of the adsorbent increased until a balance point was reached. When it reached the equilibrium point, the surface of the adsorbent was fully covered by the adsorbed dyestuff and the adsorbent reached its saturation point so that the adsorbent cannot adsorb the dye again.

3.2 Effect of Mass of The Adsorbent and Contact Time on Cu Removal Efficiency with Banana Peel and Water Hyacinth Leaves as Adsorbents

At this stage, the experiments were done with variable mass of the adsorbents (5, 10, and 15 grams), contact times (30, 60, and 90 minutes), and constant initial copper concentration (3.532 mg/L). For mixing speed parameter, the optimum is selected. From the previous experiment, it was observed that the optimum mixing speed for banana peel adsorbent was 100 rpm, while for water hyacinth leaves adsorbent was 200 rpm. The experimental results of this stage are presented in Fig. 2.

As illustrated in Fig. 2, a maximum removal of 99.55% was observed at mass of the adsorbent of 5 grams and contact time of 30 minutes using banana peel adsorbent, while using water hyacinth leaves adsorbent, a maximum removal of 98.39% was observed at mass of the adsorbent of 15 grams and contact time of 60 minutes. Therefore, the use of 5 grams banana peel adsorbent and 15 grams water hyacinth leaves adsorbent was justified for economical purposes. The increase in percent removal of adsorbate ions with increase in the adsorbent dose could be attributed to greater availability of adsorption sites. At equilibrium, the percent removal became...
constant probably because of the saturation of the available adsorption sites [18]. Increase in the adsorbent dosage enhanced the availability of more active sites for the adsorption, thus making easier penetration of metal ions to the adsorption sites [15]. At higher adsorbent dose, there are not enough Cu in the solution to occupy the active sites and hence the adsorption tends to become constant [19]. In research conducted by Thuan et al. [20], optimizing the adsorption of heavy metal ions onto banana peel derived activated carbon (AC) using response surface methodology (RSM) involving central composite design (CCD) was carried out. The maximum adsorption capacity appeared to follow the order: \( \text{Cu}^{2+} \) (14.3 mg/g) < \( \text{Ni}^{2+} \) (27.4 mg/g) < \( \text{Pb}^{2+} \) (34.5 mg/g) that agreed well with the verification experiments, revealing the reliability and suitability of the optimization approach. Mahmood et al. [21] reported that the water hyacinth plant could be able to remove metal ions like chromium, zinc and copper from the textile effluent collected from Lahore district, Pakistan. The feasibility of water hyacinth to treat wastewater from five textile effluent samples was investigated for a period of 96 h and it was observed that the water hyacinth containing textile effluent wastewaters have the potential to reduce a maximum of 94.78% reduction in chromium, 96.88% in zinc and 94.44% reduction in copper. Sarkan et al. [8] reported that after filtration, the effluent showed that the permissible limit of investigated metals. Adsorbent capacity of water hyacinth shoot powder for Cr and Cu ion was found to be 99.98% and 99.96% for standard solution (SS) and 98.83% and 99.59% for tannery effluent (TE), respectively. From some of these studies, it can be concluded that banana peel and water hyacinth leaves can be used as an adsorbent to remove Cu metal.

3.3 Adsorption Isotherm

Several models have been used to describe the experimental data of adsorption isotherm. The Freundlich and Langmuir models are the most frequently employed models [15]. The Freundlich isotherm model explains the interaction between adsorbate molecules and adsorbents with multilayer adsorption on heterogeneous surfaces. A higher value of \( n \) (\( n > 1 \)) indicates favorable adsorption, whereas \( n < 1 \) represents poor adsorption characteristics [15]. Freundlich adsorption isotherm can be seen in Fig. 3. As it is shown in Fig. 3., the value of \( n = -0.97 \) suggests poor adsorption characteristics using banana peel adsorbent, while \( n = 2.73 \) suggests favorable adsorption using water hyacinth leaves adsorbent. The situation \( n > 1 \) is most common and may be due to a distribution of surface sites or any factor that cause a decrease in adsorbent-adsorbate interaction with increasing surface density [6]. The isotherm data did not fit the Freundlich model (\( R^2 = 0.3827 \)) using banana peel adsorbent, while isotherm data fit the Freundlich model (\( R^2 = 0.8451 \)) using hyacinth leaves adsorbent. The Langmuir isotherm suggests a monolayer adsorption on a homogeneous surface, and there was no interaction between adsorbed species [3]. Langmuir adsorption isotherm can be seen in Fig. 4. The \( R^2 \) values for Langmuir isotherm model (Fig. 4) were closer to unity (0.8794 using banana peel and 0.9499 using hyacinth leaves), which indicated that Langmuir isotherm model was obeyed. This led to the conclusion that the adsorption of Cu onto the adsorbent was homogeneous and multilayer in nature. It has been suggested that for favorable adsorption the values of the dimensionless separation factor, \( R_L \) should be 0 < \( R_L < 1 \). \( R_L = 1 \) represents linear adsorption, while \( R_L < 1 \) is for favorable adsorption, while \( R_L > 1 \) represents unfavorable adsorption [6]. In this case, the value of \( R_L \) was found to be 0.013, suggesting that the adsorption process was favorable adsorption using banana peel adsorbent, while using water hyacinth leaves adsorbent, the value of \( R_L \) was found to be 0.019, suggesting that the adsorption process was favorable adsorption. The outcome values parameters \( K_f, n, K_L, b, R^2 \) for all the experiments with initial concentration 3.532 mg/l for maximum removal of copper (Cu) are presented in Table 1. As can be observed, experimental data were better fitted to the Langmuir equation than to the Freundlich equation, and therefore it is more suitable for the analysis of kinetics. Consequently, the sorption process of copper on banana peel and hyacinth leaves follows the Langmuir isotherm model, where the metal ions are taken up independently on a single type of binding site in such a way that the uptake of the first metal ion does not affect the sorption of the next ion [14].

**TABLE 1**

<table>
<thead>
<tr>
<th>Isotherm</th>
<th>Banana Peel Adsorbent</th>
<th>Hyacinth Leaves Adsorbent</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Langmuir</strong></td>
<td>25.972x-1.1664</td>
<td>0.9499</td>
</tr>
<tr>
<td><strong>Freundlich</strong></td>
<td>5.761x+0.5501</td>
<td>0.8794</td>
</tr>
<tr>
<td><strong>R^2</strong></td>
<td>0.5499</td>
<td>0.8794</td>
</tr>
<tr>
<td><strong>K_f</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>n</strong></td>
<td>-0.97</td>
<td></td>
</tr>
<tr>
<td><strong>K_L</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>b</strong></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

![Fig. 3. Freundlich Adsorption Isotherm of Banana Peel and Water Hyacinth Leaves Adsorbents](image1)

![Fig. 4. Langmuir Adsorption Isotherm of Banana Peel and Water Hyacinth Leaves Adsorbents](image2)
3.4 Water Quality Analysis

Water quality refers to the chemical, physical, biological, and radiological characteristics of water. In this case, the content of heavy metal Cu in wastewater after the adsorption process was tested as an indicator of the performance of the adsorption process using banana peel and water hyacinth leaves as adsorbents. Water quality after the adsorption process showed in Table 2.

<table>
<thead>
<tr>
<th>TABLE 2</th>
<th>WATER QUALITY AFTER THE ADSORPTION PROCESS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Contact Time (min)</td>
<td>Initial Conc. (mg/l)</td>
</tr>
<tr>
<td></td>
<td>Mass of Adsorbent</td>
</tr>
<tr>
<td>30</td>
<td>0.081</td>
</tr>
<tr>
<td>60</td>
<td>5</td>
</tr>
<tr>
<td>90</td>
<td>0.073</td>
</tr>
<tr>
<td>30</td>
<td>0.071</td>
</tr>
<tr>
<td>60</td>
<td>3.53</td>
</tr>
<tr>
<td>90</td>
<td>0.071</td>
</tr>
<tr>
<td>30</td>
<td>0.015</td>
</tr>
<tr>
<td>60</td>
<td>15</td>
</tr>
<tr>
<td>90</td>
<td>0.110</td>
</tr>
</tbody>
</table>

The quality of water produced after the adsorption process using the adsorbents of banana peel and water hyacinth leaves has met the quality standard according to the Regulation of the Minister of Environment of the Republic Indonesia Number 5 of 2014 concerning quality standards for wastewater in the paint industry, that heavy metal Cu concentration limit is 0.8 mg/L. Reduction of copper concentration occurred significantly which was very low comparing with the quality standards, whether in variations of contact time and of weight of adsorbent. The lowest effluent concentration was reach at contact time of 60 minutes, both for banana peel and water hyacinth leaves, but in difference weight of adsorbent. From these results it can be said that adsorption process using banana peel and water hyacinth leaves in this research deserve to be applied as a waste water treatment plant.

4 Conclusion

The research showed that banana peel waste and water hyacinth leaves were proved could be utilized as biosorbent for Cu with very good efficiency (more than 98%) in small amount (5-15 grams), in a quite short time (30-60 minutes) and the effluent quality meet the requirement of allowance quality standard. Mixing of speed influence the adsorption process in removing Cu heavy metal concentration, the optimum mixing speed for banana peel adsorbent was 100 rpm, while for hyacinth leaves was 200 rpm. Weight of adsorbent gives impact as well, that the use of 5 grams banana peel adsorbent and 15 grams water hyacinth leaves adsorbent were justified for economical purposes. The optimum contact time for both adsorbent was 60 minutes. Both the adsorbent fit the Langmuir isotherm model of adsorption. It could be concluded that the organic waste like banana peel and water hyacinth were promising materials to be used as biosorbents and solving the heavy metal pollution in environment. In future research, it is suggested to do the experiment in pilot plant scale.

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References


