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Effectiveness of Peanut Shell Activated Carbon as an Adsorbent for Co(II) and Cu(II) Metals in Waste Water Using a Continuous System

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ABSTRACT

This study aims to investigate the characteristics and performance of physic-chemically activated peanut shell adsorbent in reducing Co(II) and Cu(II) metal concentrations from textile industry wastewater. The activation process involved heating at 450°C and immersion in 0.5 M NaOH. Adsorbent characterization was conducted according to SNI 06-3730-1995 and analyzed using SEM-EDX. Adsorption was carried out in a continuous column system with variations in flow rate, adsorbent mass, and initial concentrations of Co(II) and Cu(II). The inactivated adsorbent had a moisture content of 3.11%, ash content of 5.54%, volatile matter of 64.01%, carbon content of 85.43%, and iodine adsorption of 261.68 mg/g. After activation, these values changed to 3.99%, 7.45%, 53.88%, 74.50%, and 355.32 mg/g, respectively. Optimum adsorption of Co(II) occurred at a flow rate of 0.0358 mL/s and 0.9 g adsorbent mass, while for Cu(II), the optimum was at 0.1282 mL/s and 0.9 g. The optimal volume ratio for both metals was 1:2 (v/v). Adsorption efficiency reached 78.34% for Co(II) and 99.98% for Cu(II), with adsorption capacities of 7.95 mg/g and 9.73 mg/g, respectively.

Keyword: : adsorption, activated carbon, metal, peanut shell, AAS

1. INTRODUCTION

The rapid development of the textile industry in Indonesia not only provides benefits for human life, but can also have a negative impact on the environment. This is caused by the waste produced, one of which is dye waste resulting from the textile dyeing process (Nurlaili, Kurniasari, & Ratnani, 2017). Heavy metal waste in the textile industry mostly comes from the dyeing process in the finishing unit. Heavy metals produced include lead (Pb), cadmium (Cd), arsenic (As), zinc (Zn), chromium (Cr), cobalt (Co), and copper (Cu) (Komarawidjaja, 2017).

Copper (Cu) is a heavy metal that is harmful to the environment. High concentrations of Cu can be harmful to aquatic life and can accumulate in the body, disrupting metabolic processes and causing toxicity (Mayangsari et al., 2019). Cobalt (Co) is also a metal known to pollute ecosystems, but it is essential for life. Cobalt poisoning in humans can cause cardiomyopathy and harm the heart muscle (Purbalisa & Dewi, 2019).

Numerous efforts have been made to address heavy metal pollution in wastewater, including ion exchange methods, membrane separation, coagulation, and adsorption. Adsorption is a frequently used method due to its low operating costs and relatively efficient process (Wang et al.,

2019). This is consistent with research by Albatrni, Qiblawey, & El-Naas (2021), which found that membrane separation requires higher additional costs than adsorption.

Using peanut shells as an adsorbent is a more environmentally friendly and economical option, as they are sourced from nature and can reduce agricultural waste (Masrofah, 2017). Furthermore, the high cellulose content in peanut shells can enhance adsorption capacity due to the presence of negative sites on the surface. Cellulose contains active sites, namely hydroxyl groups, that can form chemical reactions and bind cationic and anionic compounds (Irdhawati, Andini, & Arsa, 2016).

This study will investigate the ability of peanut shell activated charcoal to reduce Co(II) and Cu(II) metal levels in textile industrial wastewater using a continuous system analyzed using AAS. Both metals are commonly found in industrial wastewater and have the potential to impact environmental pollution. This research is expected to be an environmentally friendly alternative in addressing heavy metal pollution such as Co(II) and Cu(II), as well as providing a real contribution to the development of natural-based wastewater treatment technology.

2. RESEARCH METHOD

2.1 Peanut Shell Adsorbent Preparation

Peanut shells cut into small pieces are cleaned of impurities with running water, then dried in the sun and roasted until black. The cleaned and dried peanut shells are carbonized at 450°C for 5 minutes. The resulting charcoal is ground with a mortar and sieved through a 60 mesh sieve.

2.2 Peanut Shell Adsorbent Activation

Peanut shell powder was placed in a beaker, then 500 mL of 0.5 M NaOH was added and stirred at a constant speed for several hours, allowing for a 24-hour. The solution was then filtered until the pH reached neutral. The neutralized adsorbent was dried in an oven at 105°C for 1.5 hours. The characteristics were then determined according to SNI 06-3730-1995, including moisture content, ash content, volatile matter content, carbon content, and iodine adsorption capacity.

2.3 Artificial Waste Preparation

A 1.1015 grams and 0.2203 grams of $CoCl_2$ were placed in beakers and dissolved in distilled water. The Co(II) solution was placed in 500 mL and 100 mL volumetric flasks, respectively, and distilled water was added to the mark. 1.2558 grams of $CuSO_4$ were placed in beakers and dissolved in distilled water. The Cu(II) solution was placed in 500 mL volumetric flasks and distilled water was added to the mark. Artificial waste was prepared from a mixture of 1000 ppm Cu(II) and 1000 ppm Co(II) solutions, each containing 300 mL of each solution, in various ratios of 1:1, 1:2, and 1:3 (v/v), with the Cu(II) solution as a fixed ratio.

2.4 Determining Optimum Conditions for Co(II) and Cu(II) Adsorption

The column was cleaned using running water until it was free of impurities. 0.5 grams of cleaned glass wool was placed at the bottom of the column and compacted, followed by 0.3 grams of adsorbent. Before introducing the artificial waste, the column was first flushed with distilled water. The titrant reservoir was filled with 30 mL of artificial waste at a 1:1 (v/v) ratio, then slowly flowed continuously from the top to the bottom from the waste reservoir. The valve on the column was fully opened to vary the fast flow rate. This step was repeated with variations in mass of 0.6 grams, 0.9 grams, and slow flow rates. The filtrate obtained was collected in a sample bottle and analyzed using AAS to determine the concentration after adsorption.

3. RESULTS AND ANALYSIS

3.1 Peanut Shell Adsorbent

The production of peanut shells involves several stages. The first stage is dehydration, which begins with washing the chopped peanut shells. They are then dried in the sun until completely dry, and then roasted until they turn black. The second stage is carbonization, which aims to decompose cellulose into carbon through heating. The use of high temperatures in the carbonization process causes the evaporation of water and volatile compounds from the material, resulting in weight loss. In this study, the peanut shells were carbonized at 450°C for 5 minutes using a furnace. The carbonized peanut shells were ground using a 60 mesh sieve to reduce their size. This step aims to increase the surface area of the peanut shells, thereby increasing their adsorption capacity.

The third stage is peanut shell activation, which aims to dissolve various impurities within the pores, such as inorganic minerals, thereby expanding the cavity volume or pore system of the adsorbent and increasing its adsorption capacity (Nurhasni, Mar'af, & Hendrawati, 2018). In this study, the activation process was carried out physicochemically by heating during carbonization and immersing it in 500 mL of 0.5 M NaOH for 24 hours in a beaker. During the activation process, the beaker was covered with aluminum foil to prevent unwanted contamination. The activated charcoal was then washed with distilled water to neutralize its pH, remove impurities after the activation process, and remove any remaining NaOH remaining in the activated charcoal. Washing with distilled water was repeated until the solution reached a neutral pH, which was tested using a pH meter (Pratiwi & Setiorini, 2023).

The activated charcoal was then dried in an oven at 105°C for 1.5 hours. Drying the activated charcoal removed the water content after the activation and neutralization processes. The resulting activated charcoal was stored in a desiccator to maintain its dryness. Next, the peanut shells were ground again using a 60 mesh sieve to achieve uniform particle size.

3.2 Characterization of Peanut Shell Adsorbent

The peanut shell adsorbent formed was characterized to determine the quality of the adsorbent by referring to SNI 06-3730-1995. The results of the characteristics of the peanut shell adsorbent before and after activation are shown in Table 1.

Table 1. Characterization of Peanut Shell Adsorbent

Parameters	Adsorbent Before Activation	Adsorbent After Activation	SNI 06-3730-1995
Water Content (%)	3,11	3,99	Maks. 15
Ash Content (%)	5,54	7,45	Maks. 10
Volatile Matter Content (%)	64,01	53,88	Maks. 25
Carbon Content (%)	85,43	74,50	Min. 65
Adsorption Capacity for Iodine (mg/g)	261,68	355,32	Min. 750

3.2.1 Water Content

Water content testing is conducted to determine the amount of water contained in the peanut shell adsorbent after carbonization and chemical activation, both chemically bound and due to external factors such as weather, grain size, and the filtration process. It also aims to determine the hygroscopic properties of activated carbon. Moisture content testing is conducted before and after activation in triplicate, in accordance with SNI 06-3730-1995. Moisture content testing is performed at 115°C for 3 hours.

Table 1 shows an increase in water content after activation. This increase in water content is due to the hygroscopic nature of activated carbon, which allows it to absorb water vapor from the

air. This occurs because its structure consists of six carbon atoms, forming a hexagonal lattice that allows water vapor to be trapped within it (Susmanto et al., 2020).

3.2.2 Ash Content

Ash content testing was conducted in triplicate in accordance with SNI 06-3730-1995, by heating the samples at 800°C for 2 hours, both before and after activation. Ash content determination was performed to determine the mineral residues left over during combustion. This means that some minerals were lost during carbonization, while others remained in the peanut shells (Tasanif, Isa, & Kunusa, 2020). Ash is the result of the degradation of inorganic compounds or minerals that occur at high temperatures. Activated charcoal made from natural materials not only contains carbon compounds but also various minerals. The ash content in activated charcoal indicates its mineral content.

The increase in ash content after activation occurs due to the formation of mineral salts during the carbonization process, which then forms fine particles from these mineral salts due to the mineral content in the starting material for activated carbon. Furthermore, activation time and activation materials are factors. The longer the activation time, the more open the pores of the activated carbon, which can increase the surface area of the activated carbon (Sa'diyah & Lusiani, 2022).

3.2.3 Volatile Matter Content

Determining volatile matter levels aims to determine the content of compounds that have not evaporated during the carbonization process but will evaporate at 950°C. Measuring volatile matter levels is important for identifying the reactions that occur during the activation process between carbon and water vapor, which form volatile compounds such as CO, CO2, CH4, and H2 (Imani, Sukwiwa, & Febrina, 2021).

The volatile content in this study did not meet SNI 06-3730-1995, which states that volatile content should not exceed 25%. Volatile content is influenced by time and temperature during the carbonization process. The longer the process lasts and the higher the temperature used, the more volatile substances will be released. This means that the higher the carbonization temperature, the lower the volatile content produced (Meilianti, 2017).

3.2.4 Carbon Content

The carbon content value is the amount of carbon that does not evaporate during high-temperature heating or when determining volatile matter levels. Determining the bound carbon content aims to determine the carbon content after the carbonization and activation processes. If the charcoal contains a high bound carbon content, then the pure activated carbon content in the activated charcoal is high. Conversely, if the carbon content in the raw material is low, then the pure activated carbon content in the resulting activated charcoal will also be low. The higher the bound carbon content, the better the activated charcoal's ability to adsorb heavy metals (Rahman et al., 2018). The decrease in carbon content after activation was caused by the high ash content measured in the sample after activation and the high volatile content which tended to exceed the provisions of SNI 06-3730-1995.

3.2.5 Adsorption Capacity for Iodine

The iodine adsorption capacity for the adsorbent tested before activation (261.6805 mg/g) and after activation (355.3200 mg/g) was lower than SNI 06-3730-1995. In other words, the iodine adsorption capacity for the adsorbent before and after activation did not meet the SNI minimum limit of 750 mg/g, indicating the low quality of the adsorbent. This could be due to imperfect charcoal production and activation processes. A characteristic of perfect activated charcoal is an increase in the surface area or pores from 2 m2/g in the charcoal to 300-2000 m2/g in the activated carbon.

Another possibility is the presence of substances other than carbon, such as hydrocarbon groups and ash (Lubis, Soni, & Fauziah, 2021).

3.3 Morphology Peanut Shell Adsorbent

The results of atomic analysis on the morphology of peanut shell adsorbent using SEM-EDX showed that the peanut shell adsorbent before activation contained atomic components including carbon (C) 85.43%, oxygen (O) 14.03%, and potassium (K) 0.54%. The main components detected were carbon and oxygen. The components of the peanut shell adsorbent after activation experienced changes in the atomic components on the surface of the adsorbent. The atomic components of the adsorbent after activation included carbon (C) 74.50%, oxygen (O) 22.96%, sodium (Na) 1.77%, magnesium (Mg) 0.39%, and calcium (Ca) 0.65%. The main components detected were carbon and oxygen.

The results of SEM-EDX analysis on the adsorbent before activation and after activation are shown in Figure 1 and Figure 2 at 5,000× and 10,000× magnification. The pores formed in the adsorbent before activation as a whole are included in the macropore structure (ø > 50 nm) with a diameter range of 194.2 nm – 1,102,000 nm. After the adsorbent is activated with base treatment, there is a shift in the distribution of pore sizes towards a wider direction to 674.6 nm – 1,347,000 nm which is included in the macropore structure. The widening of the pore size is due to the loss of impurities that fill the pore system due to activation (Fisli, Safitri, & Deswita, 2018).

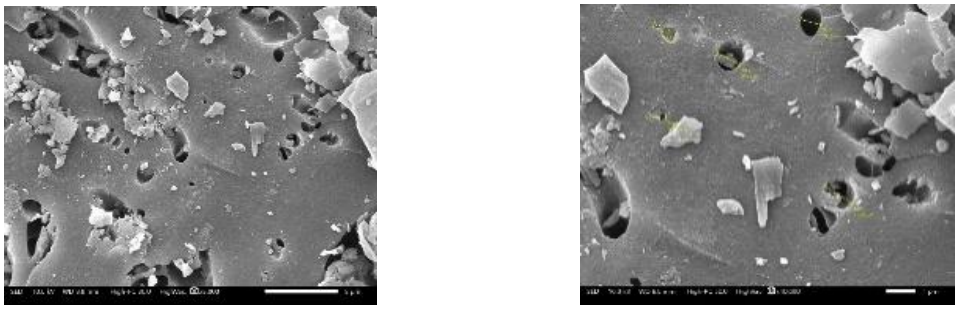

Figure 1. Adsorbent Morphology Before Activation 5,000× and 10,000×

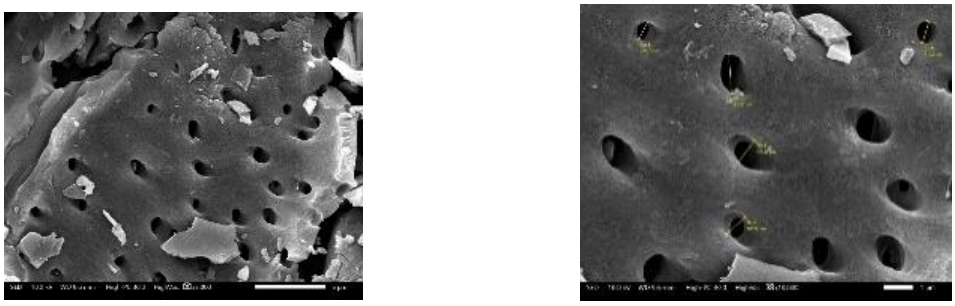

Figure 2. Adsorbent Morphology After Activation 5,000× and 10,000×

The results of SEM-EDX analysis of the adsorbent before activation showed that the charcoal surface was still closed or blocked. This is caused by the presence of residual impurities such as organic compounds in the form of minerals left on the pore surface after the carbonization process. Meanwhile, on the surface of the adsorbent after activation, the pores of the activated charcoal become open because the remaining impurities and organic compounds that previously covered the pore surface have been removed by the 0.5 M NaOH activator during the activation process. The activation process encourages the opening of the pores of the activated charcoal by breaking hydrocarbon bonds, thereby increasing its adsorption capacity (Maylani, Ismiyati, & Yustinah, 2023).

3.4 Determination of Optimum Conditions

The results of the adsorption of Co(II) and Cu(II) metals using activated charcoal from peanut shells applied to a continuous column system with variations in flow rate, adsorbent mass, and concentration of Co(II) and Cu(II) metals.The results of determining the optimum conditions were obtained at the highest adsorption efficiency values for Co(II) and Cu(II) metals. The effect of variations in mass, flow rate, and concentration of Co(II) and Cu(II) metals on adsorption efficiency can be seen in Figure 3 and Figure 4.

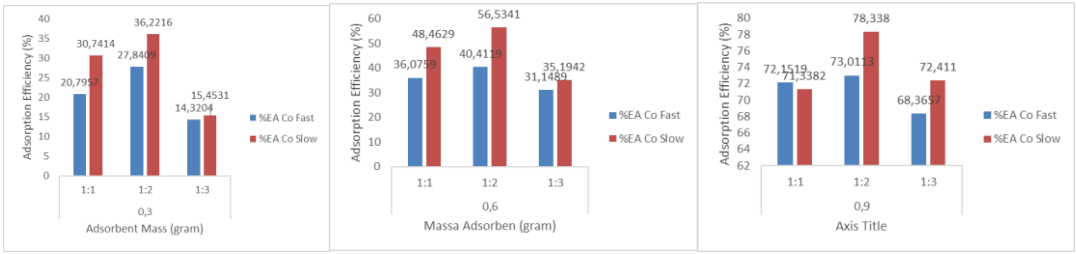

Figure 3. Relationship between Mass Variation, Flow Rate, and Metal Concentration on Co(II) Adsorption Efficiency

Figure 3 shows the highest Co(II) metal adsorption efficiency at a flow rate of 0.0358 mL/s with an adsorption efficiency of 78.3380%. The highest Co(II) metal adsorption efficiency at slow flow rates is related to the contact time of the adsorbent with the artificial waste solution. The longer the artificial waste solution is in the column and in contact with the adsorbent, the more effective the reduction in heavy metal levels in the artificial waste becomes. This creates more opportunities for bonding to occur with the active groups of the adsorbent.

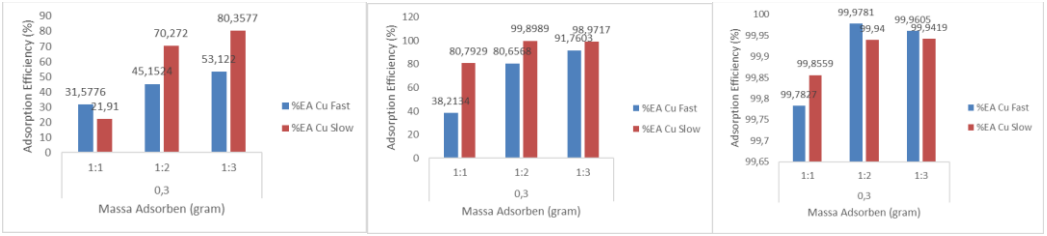

Figure 4. Relationship between Mass Variation, Flow Rate, and Metal Concentration on Cu(II) Adsorption Efficiency

Figure 4 shows that the optimum conditions for Cu(II) metal were at a flow rate of 0.1282 mL/s with an adsorption efficiency of 99.9781%. The adsorption efficiency of Cu(II) metal was greatest at a fast flow rate. This may occur because copper tends to have a higher affinity for peanut shell activated charcoal, making activated charcoal more effective in adsorbing copper metal even within a short contact time (Putri & Hardiansyah, 2022). The optimum mass of both was obtained at 0.9 grams with adsorption efficiencies of Co(II) and Cu(II) metals of 78.3380% and 99.9781%, respectively. This occurs because the increase in the number of adsorbents results in an increase in the surface area and active sites on the adsorbent surface. The increasing amount of adsorbent means that more Co(II) and Cu(II) metals are adsorbed (Suwantiningsih, Khambali, & Narwati, 2020).

The optimum condition for both is at a volume ratio of 1:2 (v/v), because when the concentration variation of Co(II) > Cu(II) provides flexibility for Co(II) to control the mass transfer process, thereby increasing the opportunity for Co(II) to contact or collide with the active side of the adsorbent. This also occurs in the research of Indah, Helard, & Yedriana (2016) which states that the greater the number of Fe ions in a solution in the adsorption process causes the greater flexibility for Fe metal ions to control the mass transfer process from the adsorbate to the adsorbent.

The difference in the number of adsorbed ions can also be determined based on the radius of the metal ion. Seen from the periodic system of elements, the ion radius tends to be larger based on the group from top to bottom, indicating an increase in the electron shell, while in the period there is a decrease from left to right. From the periodic table of elements, it can be seen that Co and Cu are in the same period with atomic numbers Co = 27 and Cu = 29. Therefore, it can be stated that the radius of Co is larger than the radius of Cu, cobalt has an ionic radius of 1.52 Å while copper has an ionic radius of 1.45 Å. The smaller the ionic radius of a metal, the greater the attraction of the nucleus to the ion, which causes the metal to quickly bind to the active surface of the adsorbent (Kristianingrum et al., 2020).

4. CONCLUSION

Based on this research, it can be concluded that peanut shells can be used as heavy metal adsorbents, especially Co and Cu. The adsorption capacity and efficiency for Co(II) metal were 7.9495 mg/g and 78.3380%, respectively. The adsorption capacity and efficiency for Cu(II) metal were 9.7266 mg/g and 99.9781%, respectively.

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