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(RESEARCH ARTICLE)

Evaluating the effectiveness of heavy metals removal from mine wastewater using agricultural waste materials

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Abstract

Polluted water bodies pose a danger to both aquatic and terrestrial lives, and recent development in the mining centre has escalated this pollution through the disposal of its industrial wastes. The objective was to investigate the potential of four different agricultural waste materials in eliminating the heavy metals from mine wastewater. The results show that the best performance in terms of adsorption capacity and removal efficiency was observed with coconut husk-activated carbon. The Arsenic (As) concentration decreased from 3.185 mg/kg to 1.570 mg/kg, and the Mercury (Hg) content decreased from 0.0307 mg/kg to 0.0064 mg/kg. The corresponding removal efficiencies were 51% for Arsenic and 79% for Mercury after 120 minutes of contact time at a speed of 250 rpm. The results suggest that coconut husk-activated carbon exhibited superior adsorption capacity and removal efficiency compared to the other agricultural waste materials (maize cob, rice husk, and sawdust). This indicates that the natural properties of coconut husk are more effective in removing heavy metals from mine wastewater.

Keywords: Heavy metals; Mine wastewater; Maize cob; Rice Husk; Coconut Husk; Sawdust

1. Introduction

Wastewater discharge from industries, agricultural activities, municipal wastewater, environmental changes, and global warming are the main causes of water contamination. The concentrations of heavy metals, dyes, and microbes pose serious risks to the environment, aquatic life, and human health. Water is used for a variety of things in our daily lives, including domestic, agricultural, and industrial needs. In essence, water is necessary for human survival on Earth. Its ineffective management is the problem, despite its significance.

Heavy metal pollution harms the world's ecosystems and human health, and it tends to become a serious environmental issue when it occurs in water sources, especially in mine wastewater. Heavy metal contamination in mine wastewater discharge poses serious threats to nearby aquatic life, water supplies, and perhaps even humans through the food chain. Efficient and sustainable solutions for heavy metal removal from mine wastewater are vital to reduce the negative effects on the ecosystem and preserve water quality.

Agricultural waste has recently attracted more attention as a viable adsorbent material for the removal of heavy metals due to its ready availability, low cost, and eco-friendly makeup. Agricultural waste, including sawdust, rice husk, coconut husk, and maize cob, has demonstrated remarkable potential as a precursor for the synthesis of activated carbon, which is capable of efficiently adsorbing heavy metals from aqueous solutions. Rice husk and activated carbon silica were employed as bio adsorbents for wastewater treatment [1]. According to their findings, pH 6 offered the optimal conditions for the removal of lead (Pb) and cadmium (Cd), and a higher metal starting concentration decreased the

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removal effectiveness of the bio adsorbents. A maximum metal removal of 81% and 98% for Pb and 88% and 100% for Cd, respectively, was achieved when rice husk and activated carbon-silica were used at pH 6.

Vunain et al. [2] investigated the chromium ion adsorption from tannery effluents onto activated carbon made from potato peel and rice husk. The highest Cr (VI) uptake, and adsorption obtained occurs at pH 2.0, with chromium removal efficiencies of 99.88% and 99.52% for rice husk activated carbon and potato peel activated carbon, respectively. Chromium (VI) ions from wastewater can be removed using activated carbon made from potato peel and rice husk.

Patabandige et al. [3] investigated the use of rice husk-activated carbon for the adsorption of synthetically coloured wastewater. According to their findings, the batch adsorption performed at its optimum in an acidic medium with a pH level of 2, an adsorbent dosage of 13 mg/l, and an agitation speed of 100 rpm, with the maximal dye removal occurring after 10 minutes. With a final dye concentration of 10.8 mg/l, the absorbent's maximum adsorption capacity was discovered to be 2.0 mg/g.

Cheng et al. [4] looked into the clean removal of lead and cadmium from wastewater using sustainable biochar made from poplar sawdust. At pH 5, Pb^{2+} has the highest adsorption capacity at 62.68 mg/g whereas Cd^{2+} has the lowest at 49.32 mg/g. It was also investigated how well Pb^{2+}/Cd^{2+} adsorbed in various water systems. With a contribution percentage of 51.76%, mineral precipitation containing Pb^{2+} is the main mechanism for Pb^{2+} elimination. At 59.36% of the total adsorption capacity, coordination with π electrons is the main mechanism for Cd^{2+} adsorption.

Ramirez et al. [5] explored the removal of Cr (VI) using activated carbon made from teakwood sawdust in an aqueous solution. The highest possible adsorption capacity of 72.46 mg g^{-1} was attained.

Al-Sareji et al. [6] investigated the use of carbonized sawdust for copper removal from water. Following the batch studies, a 150-minute maximum contact time was decided upon. The results showed that the copper concentration in the aqueous solution was reduced by around 79% by the sawdust dosage of 2 g/l.

The effectiveness of the alkaline-modified sawdust for metal removal was examined under various initial concentrations of Cu (II) and Zn (II) from model solutions. Copper adsorption efficiency levels of 94.3% at pH 6.8 and zinc adsorption efficiency values of 98.2% at pH 7.3 were the greatest for poplar treated by KOH. The group members discovered that for all types of sawdust, the sorption effectiveness of modified sorbents was greater than that of untreated sawdust. When zinc was removed from modified sawdust with spruce NaOH, the pH value first rose significantly (8.2) before gradually falling (7.0 for Zn (II) with spruce NaOH) [3].

The adsorption characteristics of modified maize cob activated carbon for mercury ions were investigated by Ajala et al. [7] both modified corn cob and activated carbon from corn cob have maximal adsorption capacities of 222.22 mg/g and 184.76 mg/g, respectively.

Ahmad et al. [8] looked at the competitive adsorption of Cu^{2+} and Ni^{2+} on corn cob-activated carbon as well as the differences in thermal impacts on mono and bicomponent systems. For Cu^{2+} and Ni^{2+}, the equilibrium was reached after 240 and 100 minutes, respectively. Cu^{2+} and Ni^{2+} have adsorption capabilities of 0.39 mmol/g and 0.28 mmol/g, respectively.

High-performance iron removal from aqueous solutions was explored by Patabandige et al. [3] utilizing modified activated carbon made from corn cobs and luffa sponge. After a contact time of 5 min and an initial iron concentration of 5 mg/l, the ideal pH was discovered to be 8 for all adsorbents, with maximum removal efficiencies of 89.3%, 99.1%, 79.7%, and 96.7% using corn cob-activated carbon, aluminium chloride corn cob activated carbon, luffa sponge activated carbon, and aluminium chloride luffa sponge activated carbon, respectively. With an initial iron content of 40 mg/L and an adsorbent dosage of 0.1 g/l, the highest adsorption capacities were 334.9, 366.7, 317.1, and 348.8 mg/g for corn cob activated carbon, luffa sponge activated carbon, and aluminium chloride luffa sponge activated carbon, respectively.

A low-cost adsorbent made from corn cobs was used to remove nickel (II) in a study by Mishra et al. [9] According to their findings, a batch study was performed, and variables like pH, contact time, adsorbent dose, and metal concentration were adjusted while the experiment was conducted at room temperature. According to the results, nickel could be completely removed after 100 minutes at pH 8.

Patabandige et al. [3] investigated the treatment of industrial wastewater using activated carbon made from coconut shells. According to their findings, a dose of 15 g/l and a time frame of 90 min is sufficient to remove 90% of phosphate and 97% of zinc for particle sizes of 150 µm.

Kwasi Opoku et al. [10] investigated the adsorption of some heavy metal contaminants in used lubricating oil using chemically activated carbon adsorbents produced from palm kernel and coconut shells. It was found that whereas palm kernel-activated carbons cannot remove lead metals, coconut shell-activated carbons could be used. However, the copper and iron metals cannot be removed using activated carbons made from coconut and palm kernel shells.

Siriweera and Jayathilake [11] explored the modifications of coconut waste as an adsorbent for the removal of heavy metals and dyes from wastewater. These modification processes include acid and alkali treatments, modification with surfactants, polymerization, and treatment with metal/metal chlorides as well as thermal treatments. According to them, modified coconut-based adsorbents could provide high water treatment capacities through enhanced adsorption efficiencies.

Commercial adsorbents are frequently expensive and may have negative environmental effects. Meanwhile, not many studies have been done on using local agricultural waste to remove heavy metals from the mine wastewater. Hence, to lessen or stabilise heavy metals from mine water waste, it would be beneficial to explore means of removing the heavy metal from mine water waste using local agricultural waste such as maize cobs, rice and coconut husks, and sawdust.

The objective of this study was to evaluate different agricultural wastes in the removal of heavy metals from mine wastewater.

2. Materials and method

2.1. Instruments

An open furnace was used to heat the agricultural waste. An oven was used to dry the samples. A ball mill was used to mill the samples. An orbital shaker was used to mix the samples thoroughly. A pH meter was used to measure the pH of each sample. A measuring cylinder was used to measure the quantity of distilled water. A conical flask was used during the agitation process. A ceramic mortar and pestle were used to grind the samples. An electronic balance was used to measure the weight of the samples. A 125 microns sieve was used to sieve the samples. Sodium chloride and distilled water was used during the chemical activation. An Atomic Absorption Spectrometer (AAS) was used to determine the elemental concentration of heavy metals in the sample.

2.2. Preparation of carbon

Figures 1, 2, 3, and 4 show samples of **e**ach agricultural waste (sawdust, rice husk, coconut husk, and maize cob) respectively that was turned into carbon by heating it for 10 minutes in an open furnace. To prevent carbon from turning into ash, water was sprinkled on each carbon as soon as it was withdrawn from the furnace. Before milling, each carbon was allowed to cool for an hour. In a ball mill, the carbons were milled for 50 minutes. Following grinding, the samples were dried in an oven at 100 ^{0}C for 3 hours and they were cooled to room temperature for an hour. The 125 microns were used to filter out larger particles, which increased the uniformity of the activated carbon produced. The samples were weighed after being cooled to room temperature. The maize cob weighed 115 g, the rice husk 76 g, the sawdust 61 g, and the coconut husk 26 g. Before and after filtering, 2 g and 3 g of each were used to determine the initial pH.

Figure 1 Maize cob

Figure 2 Rice Husk

Figure 3 Sawdust

Figure 4 Coconut Husk

2.3. Desorption of carbon

10 g and 6 g of sodium chloride were fully dissolved in 200 ml of distilled water after the thermal activation. 10 g of sodium chloride was placed into three conical flasks and 6 g into the fourth. The quantity of sodium chloride was calculated using Equation 1.

$$NaCl = \frac{P}{100} \times WAW \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots.(1)$$

Where P, is the percentage of NaCl based on the weight of the agricultural waste, and WAW is the weight of the agricultural waste in g.

The four solutions, which contained distilled water, carbon, and sodium chloride, were agitated for 30 minutes at a speed of 250 rpm using an orbital shaker to chemically activate the carbon, increasing its surface area and pore volume. The mixture was finally filtered and washed with distilled water [12]. The role of sodium chloride is to enlarge the pores of the thermally activated carbon, improving the sample's ability to absorb heavy metals. Distilled water is used to regulate the pH. The samples were dried in an oven at 60 ^{0}C for three hours before being allowed to cool to room temperature for an hour.

2.4. Characterization of activated carbon

The activated carbon was characterized by measuring its particle size, colour, mass, pH and percentage weight loss. The percentage weight loss (PwL) was calculated with Equation 2.

$$\text{Weight Loss \%} = \frac{\text{Initial Weight} - \text{Final Weight}}{\text{Initial Weight}} \times 100\% \quad \ldots\ldots\ldots\ldots.(2)$$

2.5. Adsorption experiment

Arsenic and mercury were among the adsorbates considered in this investigation. The experiment was carried out in four conical flasks. The experiment employed 300 ml of mine wastewater and 10 g of each adsorbent, namely rice husk, sawdust, coconut husk, and maize cob-activated carbons (particle size 125 microns). The elemental content of heavy metals in each solution was determined using an AAS analysis. The four combinations were stirred for two hours and filtered every 30 minutes.

The quantity of activated carbon (QAC) was calculated using Equation 3.

$$QAC = \frac{Quantity\ of\ heavy\ metals\ to\ be\ removed}{Adsorption\ capacity} \ldots\ldots\ldots\ldots\ldots (3)$$

The percentage removal (R) of heavy metals was calculated with Equation 4.

$$\mathrm{R\%} = \frac{\mathrm{Co} - \mathrm{Ct}}{\mathrm{Co}} \times 100 \ \ldots\ldots\ldots\ldots\ldots\ldots..(4)$$

The equilibrium adsorption capacity (EAC) of the adsorbent was calculated using Equation 5.

$$\mathrm{EAC} = \frac{(\mathrm{Co} - \mathrm{Ce})\mathrm{V}}{\mathrm{W}} \ \ldots\ldots\ldots\ldots..(5)$$

The quantity of adsorbed metal ions (qt) was calculated using Equation 6.

$$\mathrm{qt} = \frac{(\mathrm{Co} - \mathrm{Ct})\mathrm{V}}{\mathrm{W}} \ \ldots\ldots\ldots\ldots\ldots\ldots.(6)$$

where R is the removal efficiency of heavy metals, Co is the initial concentration, Ct is the final concentration of heavy metals in mg/l at the time (t), Ce is the final concentration of heavy metals in mg/l at equilibrium, and EAC is the equilibrium adsorption capacity of adsorbent in mg/kg, V is the volume of mine wastewater in ml, W is the weight of adsorbent in g and qt is the quantity of adsorbed metal ions.

2.6. Materials and equipment

An open furnace was used to heat the agricultural waste. An oven was used to dry the samples. A ball mill was used to mill the samples. An orbital shaker was used to mix the samples thoroughly. A pH meter was used to measure the pH of each sample. A measuring cylinder was used to measure the quantity of distilled water. A conical flask was used during the agitation process. A ceramic mortar and pestle were used to grind the samples. An electronic balance was used to measure the weight of the samples. A 125-micron sieve was used to sieve the samples. Sodium chloride and distilled water were used during the chemical activation.

Figure 5 Open furnace

Figure 6 Electronic Balance

Figure 7 Microns sieve

Figure 8 ceramic and Pestle

Figure 9 Conical flask

Figure 10 Sampling bottles

Figure 11 Orbital shaker

Figure 12 Oven

An Atomic Absorption Spectrometer (AAS) was used to determine the elemental concentration of heavy metals in the sample. Figure (5-12) shows the various materials and instruments used for the experiment. The experiment was performed at the University of Mines and Technology, Tarkwa (Ghana) minerals Laboratory.

3. Results and discussion

3.1. pH of adsorbents

Figure 13 shows that the pH of maize cob, rice husk, coconut husk and sawdust before activation is 9.80, 8.31, 10.32 and 9.95 respectively. Figure 14 shows the pH of maize cob, rice husk, coconut husk and sawdust after activation is 8.90, 7.14, 9.90 and 9.61 respectively. In the case of coconut husk, which has a high pH, it implies that the adsorbent material may have a higher adsorption capacity for heavy metals at alkaline conditions. Maize cob and sawdust also have relatively high pH values, suggesting the potential for a higher adsorption capacity. Rice husk has a lower pH compared to the other carbons, indicating that it may have a relatively lower adsorption capacity for heavy metals. The specific impact of pH on removal efficiency depend on the behaviour and characteristics of the heavy metals and the adsorbent material being used. But according to Al-Sareji et al. [6], the pH of the solution influences the surface charge of the adsorbent material as well as the ionization state of the heavy metals, influencing adsorption capacity and efficiency.

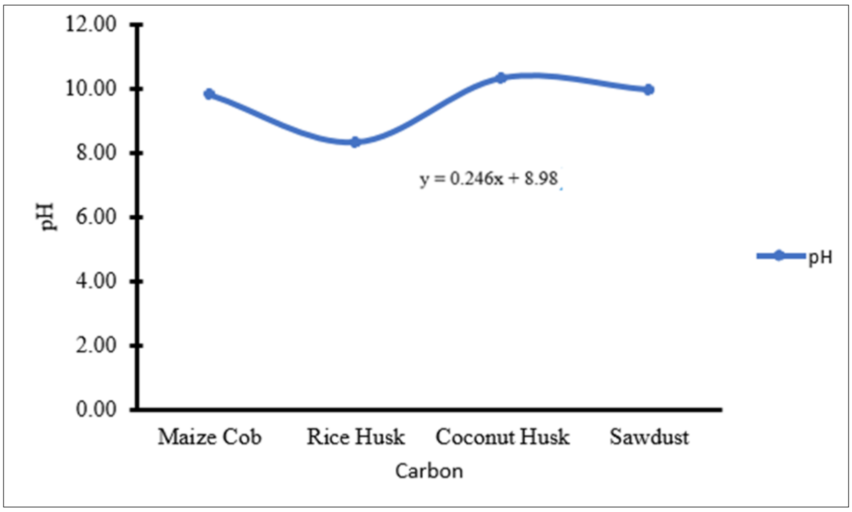

Figure 11 PH before Activation

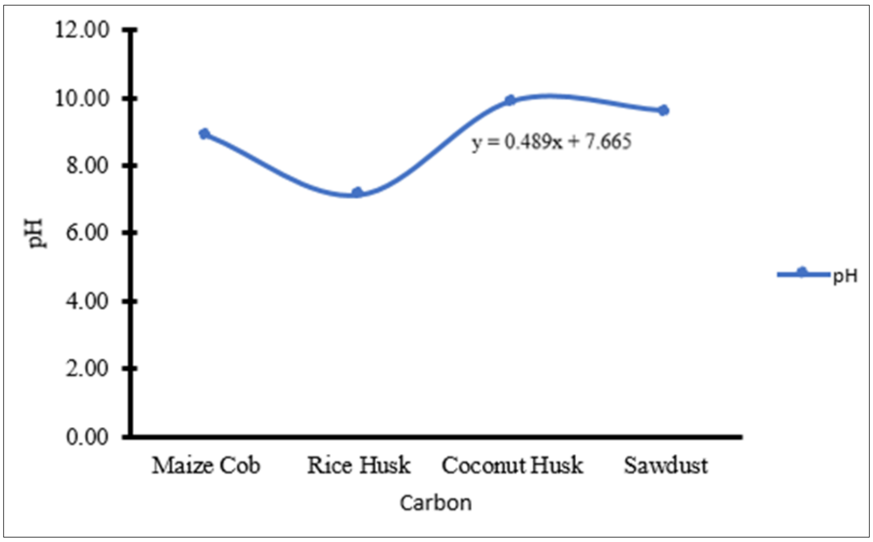

Figure 12 pH after Activation

3.2. Effect of contact time

The term "contact time" describes how long activated carbon was in direct contact with heavy metal in the mining wastewater. It is very important in figuring out the adsorption capacity and effectiveness of activated carbon for removing heavy metals. Figure 15 and 16 indicate adsorption capacity of mercury (Hg) and arsenic (As) respectively. All the samples were agitated for 120 minutes, and the agitation speed was 250 rpm.

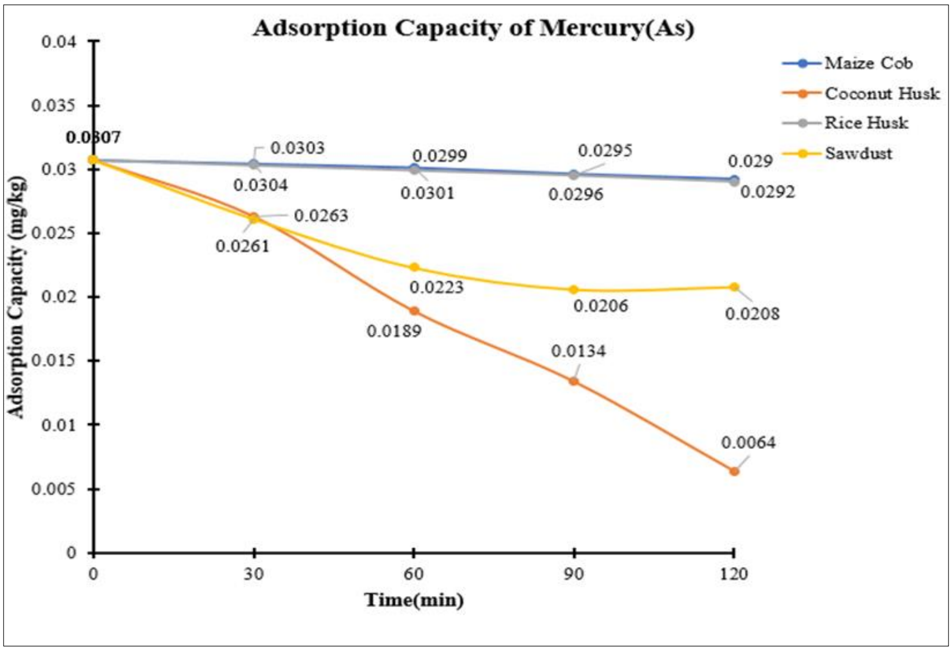

Figure 13 Adsorption capacity of Mercury (Hg)

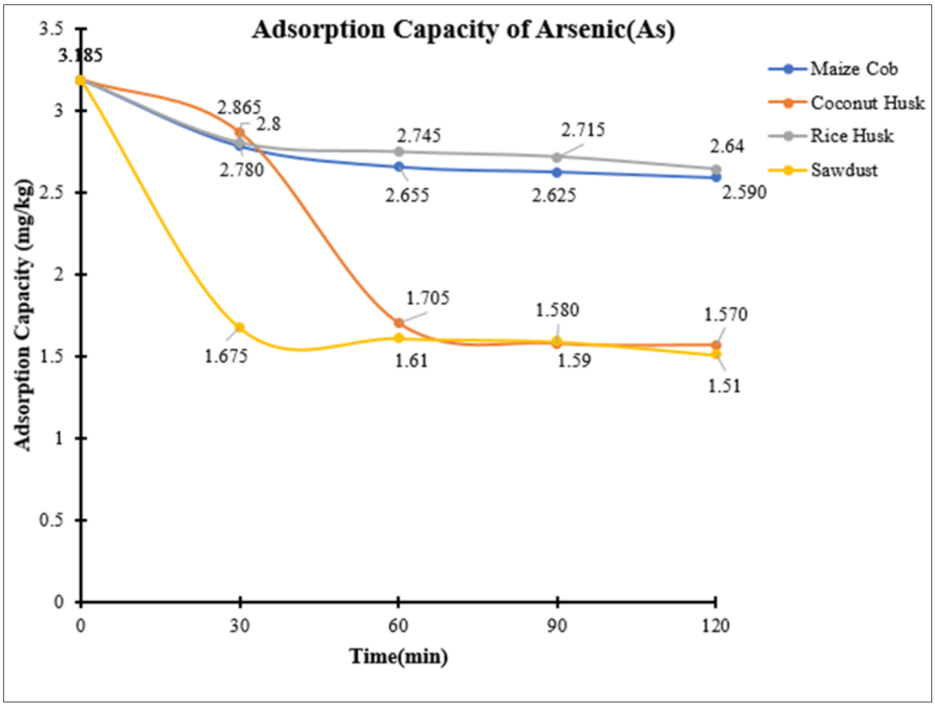

Figure 14 Adsorption capacity of Arsenic (As)

Arsenic (As) and mercury (Hg) had initial concentrations of 3.05 mg/kg and 0.0303 mg/kg, respectively, according to Fig. 15 and Fig. 16. Arsenic concentration decreased gradually from 3.05 mg/kg to 2.590 mg/kg as the samples were agitated for 120 minutes at intervals of 30 minutes with maize cob-activated carbon, while mercury concentration decreased gradually from 0.0303 mg/kg to 0.0292 mg/kg. This finding suggests that maize cob has potential as an agricultural waste-based adsorbent for the removal of heavy metals during the process of treating wastewater. It indicates that the qualities of a maize cob employed as activated carbon adsorb and lower the amounts of both arsenic and mercury in mine wastewater. Through physical and chemical adsorption mechanisms, the pores in maize cob function as microscopic traps that can catch and hold heavy metal ions. Maize cob also contains natural materials including cellulose, lignin, and hemicellulose that can aid in the adsorption of heavy metal ions. The results disagree with the study by Sanka et al. [13] that adsorption capacity of rice husk is more effective than maize cob.

From Fig. 15 and 16, as the samples were agitated with coconut husk-activated carbon for 120 minutes at intervals of 30 minutes, the Mercury content steadily declined from 0.0303 mg/kg to 0.0064 mg/kg and Arsenic concentration progressively fell from 3.05 mg/kg to 1.570 mg/kg. This indicates that the concentrations of Arsenic and Mercury in the mine wastewater were effectively reduced during the adsorption process using coconut husk-activated carbon. The reduction in the concentration of the two metals suggests that the coconut husk-activated carbon successfully adsorbed heavy metals from the mine wastewater. The effectiveness of the adsorption process can be attributed to the natural properties of the coconut husk which include; large surface area, porous structure of coconut husk and chemical composition. Ramirez et al. [5] looked at employing limestone and coconut shell carbon as adsorbents to remove Cr (III) from industrial effluent. The best adsorption capacity of Cr (III) was 0.000019 mg/g.

The initial concentrations of arsenic (As) and mercury (Hg) were 3.05 mg/kg and 0.0303 mg/kg, respectively, Arsenic levels continuously decreased from 3.05 mg/kg to 2.640 mg/kg and mercury levels steadily decreased from 0.0305 mg/kg to 0.0290 mg/kg while the samples were agitated with rice husk activated carbon for 120 minutes at intervals of 30 minutes. This suggests that the adsorption process using rice husk-activated carbon for the removal of heavy metals from mine wastewater was somewhat effective. This indicates that the rice husk-activated carbon was able to adsorb and reduce few of the concentrations of heavy metals during the 120 minutes agitating period. Rice husk has a significant surface area due to its fibrous and porous structure. The surface area and porosity of rice husk may not be as high as other materials, resulting in a lower number of adsorption capacities.

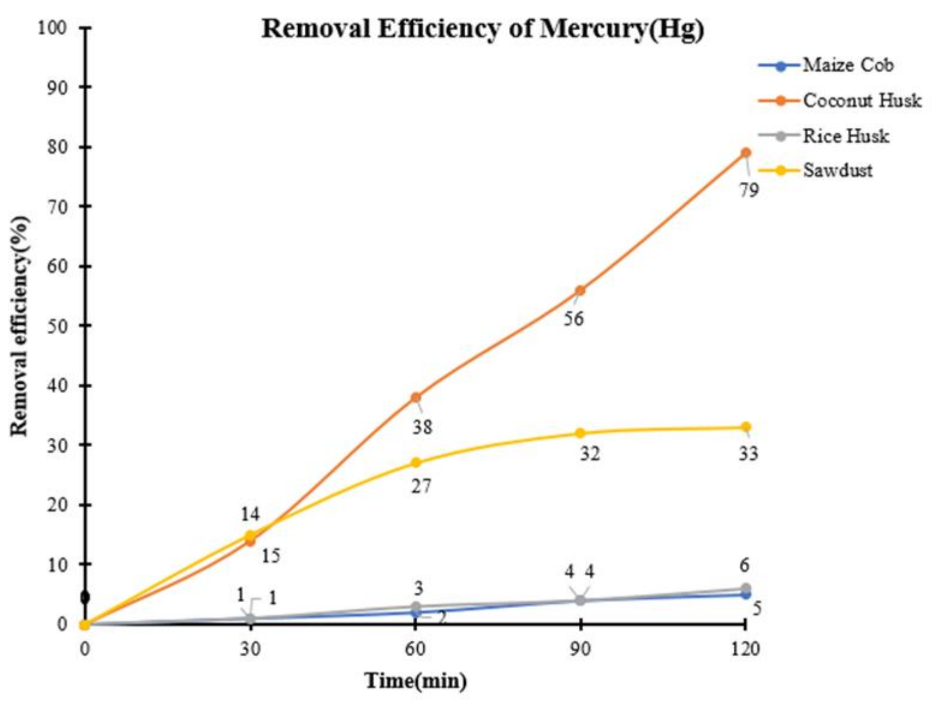

Figure 17 Removal efficiency (%) of mercury (Hg)

With the same initial concentrations of Arsenic (As) and Mercury (Hg) of using sawdust, arsenic levels progressively declined from 3.05 mg/kg to 1.510 mg/kg and mercury levels slowly decreased from 0.0303 mg/kg to 0.0208 mg/kg. The results imply that the adsorption process using sawdust-activated carbon was effective in reducing the concentrations of Arsenic and Mercury in the mine wastewater. The decrease in concentrations indicates that the activated carbon was able to adsorb and remove a significant amount of these heavy metals from the mine wastewater. The high porosity of sawdust allows for increased contact between the adsorbent and the heavy metals, enhancing the adsorption process. Ramirez et al. [5] explored the removal of Cr (VI) using activated carbon made from teakwood sawdust in an aqueous solution. The highest possible adsorption capacity of 72.46 mg g−1 was attained. Figures 17 and **18** showed that the removal efficiencies of Mercury (Hg) and Arsenic (As) at 120 minutes of contact time were 5% and 19%, respectively using maize cob-activated carbon. The results imply that the adsorption process using maize cob-activated carbon for the removal of Mercury (Hg) and Arsenic (As) from mine wastewater was not highly effective. The relatively low removal efficiencies indicate that the activated carbon within the given contact time did not successfully adsorb a significant portion of the heavy metal ions. Different heavy metals have varying affinities for different types of adsorbents, and it's possible that maize cob-activated carbon may not have a strong affinity for Arsenic (As) and Mercury (Hg). In a study conducted by Sanka et al. [13], corn husks produced the worst removal efficiencies for copper and lead.

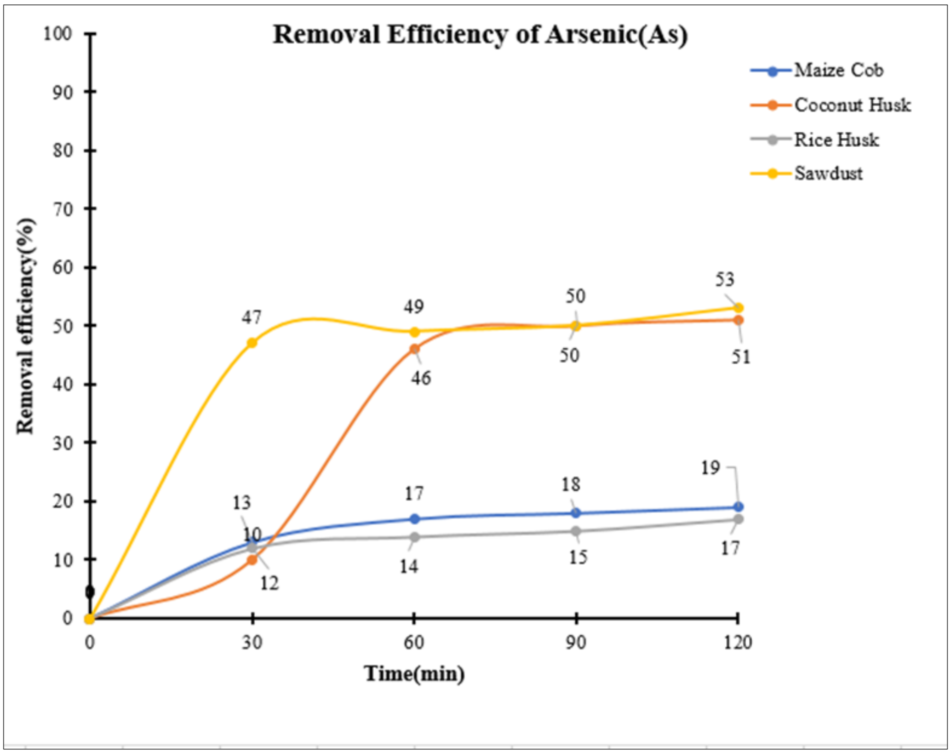

Figure 18 Removal efficiency (%) of Arsenic (As)

Mercury (Hg) and Arsenic (As) had removal efficiencies of 79% and 51% respectively, (see Fig 17 and 18), after 120 minutes of contact time using coconut husk-activated carbon the removal efficiencies suggest that a significant portion of the Arsenic and Mercury present in the mine wastewater was successfully adsorbed by the coconut husk. The results imply that the adsorption process utilizing coconut husk-activated carbon was successful in reducing the concentrations of Arsenic and Mercury in the mine wastewater. The effectiveness of the adsorption process can be attributed to the natural properties of coconut husk, which can contribute to its adsorption capacity for heavy metals. Packialakshmi et al. [14] achieved 90% removal efficiency of phosphate and 97% removal efficiency of zinc for particle sizes of 150 µm using coconut shell activated carbon.

The removal efficiency of Mercury (Hg) and Arsenic (As) using rice husk-activated had removal efficiencies of 6%, and 17% respectively (see Fig. 17 and 18), after 120 minutes of contact time using rice husk-activated carbon. The low removal efficiencies suggest that the adsorption of heavy metals using rice husk-activated carbon was not highly effective in this study. The relatively low percentage of heavy metal removal implies that a significant portion of the heavy metals remained in the mine wastewater even after the adsorption process. The natural properties of rice husk, such as its surface area and porosity might have limited adsorption capacity for Arsenic and mercury. A study was conducted by Syuhadah and Rohasliney [15] using zinc concentrations reduced to 70% using rice husk. However, removal reached 99% when coconut coir and moringa seeds were used. Meanwhile, mercury (Hg) and Arsenic (As) had removal efficiencies of 33% and 53%, respectively, after 120 minutes of contact time using sawdust-activated carbon. The relatively high removal efficiencies suggest that the adsorption process using sawdust-activated carbon was effective in reducing the concentrations of Mercury and Arsenic in the mine wastewater. However, some possible factors contributing to the moderate removal efficiencies include dosage, pH, agitation speed, and contact time. The percentages indicate the proportion of heavy metals that were successfully adsorbed and removed from the water. Kovacova et al. [16] used sawdust to remove Copper and Zinc from wastewater the removal efficiencies were 94.3% and 98.2% respectively.

The worst performance was observed with maize cob-activated carbon. Arsenic (As) concentration decreased from 3.05 mg/kg to 2.590 mg/kg, and the Mercury (Hg) concentration decreased from 0.0307 mg/kg to 0.0292 mg/kg. The removal efficiencies were relatively low, with 19% for Arsenic and 5% for Mercury.

The results suggest that coconut husk-activated carbon exhibited superior adsorption capacity and removal efficiency compared to the other agricultural waste materials (maize cob, rice husk, and sawdust). This indicates that the natural properties of coconut husk are more effective in removing heavy metals from mine wastewater.

4. Conclusions

Based on the results, it can be concluded that the use of activated carbon derived from agricultural waste shows promise for the removal of heavy metals from mine wastewater. The Mercury (Hg) content decreased from 0.0303 mg/kg to 0.0064 mg/kg. The corresponding removal efficiencies were 51% for Arsenic and 79% for Mercury, while the Arsenic (As) concentration decreased from 3.05 mg/kg to 1.570 mg/kg, however, the achieved removal efficiencies varied depending on the adsorbent material used. The pH values of the adsorbents before and after activation indicate their suitability for the adsorption process. The best performance in terms of adsorption capacity and removal efficiency was observed with coconut husk-activated carbon.

Compliance with ethical standards

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Disclosure of conflict of interest

Authors declare that there is no competing conflict of interests

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