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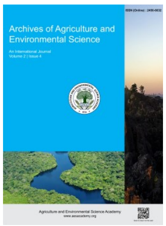

ORIGINAL RESEARCH ARTICLE

Wastewater treatment using coconut fibre ash as an adsorbent for removal of heavy metals

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ABSTRACT

The study aimed at evaluating the performance of coconut fibre ash as an alternative low-cost adsorbent to the synthetic adsorbents used in wastewater treatment. This research aims to identify the optimum condition for the adsorption process, considering the effect of particle size, adsorbent dosage, and contact time of adsorbents of coconut fibre ash in removing lead (Pb), copper (Cu), and zinc (Zn) metal ions from electroplating wastewater. The adsorbents coconut fibre ash was prepared through activation of carbon at 450° C after following proper cleaning and drying process. The experiments were conducted at varying adsorbent dosages (0.2 g, 0.6 g, and 1 g), particle size (50 to 200 microns), and contact times (40 minutes, 80 minutes, and 120 minutes). The result shows that adsorbents show less efficiency in removing Zn metal ions, which is not more than 34% in the case of 1g adsorbent dosage, particle size ranges 100-200 microns, and 120 minute contact time. The maximum removal efficiency of 95.04% and 80% was obtained at the optimum amount (1g) of adsorbent dosage for Pb and Cu respectively. In the case of contact time, it was identified that the optimum condition for maximum removal efficiency is 120 minutes with a 1g adsorbent dosage both for Pb and Cu ions. To ensure maximum removal of metal avoiding any desorption of the metal ion from the adsorbent surface, it was identified that a maximum contact time of 120 minutes should be allowed for adsorption. However, it could be concluded that adsorbents of coconut fibre ash can be used in treating wastewater facilitating good adsorption capacity in removing heavy metals, low cost and availability.

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INTRODUCTION

Water pollution caused by industrial wastewater has become a serious environmental problem (Kumar *et al*., 2019; 2021; Singh *et al*., 2021a,b). Increased industrial activities have caused the heavy metal load of many sources of water to exceed the maximum allowable limit for wastewater discharge designed to protect the environment, humans, and animals (Iqbal and Saeed, 2007). Various industrial wastewater streams can contain heavy metals such as chromium, copper, lead, zinc, nickel, mercury, etc. including waste liquids from the metal finishing, electroplating, textile, or mineral processing industries. The accumulation of these toxic heavy metals into our human body results in chronic health effects due to their presence of them in soft and hard tissues. There are at least twenty types of heavy metals that are toxic and half of these are emitted to the environment in quantities that pose risk to human health (Atif *et al*., 2009). Zinc metal is hazardous in the case of skin and eye contact, poses indigestion, and is hazardous if inhaled. Though zinc is necessary for the human body, the accumulation of the excess amount of zinc causes skin diseases, stomach cramps, vomiting, and anemia (Oyaro *et al*., 2007). Lead has adverse effects on the plant due to the inhibition of photosynthesis (Ekmekci *et al*., 2009). Exposure to a high level of lead may result in anemia, weakness, and kidney and brain damage in the human body. Copper has both toxic and chronic effects on living and aquatic organisms that can result in death. From the ecosystem's point of view, the toxicity of the heavy metal has a great threat to the aquatic environment if there is no proper treatment for the industrial effluent and wastewater before discharging them directly into the environment. So, it is important to ensure the removal of heavy metals from wastewater.

Different conventional treatment technologies applied for the removal of heavy metals are ion exchange, chemical precipitation, chemical oxidation, reduction, reverse osmosis, ultrafiltration, electrodialysis, and adsorption (Fu and Wang, 2011). For example, as a cost-effective method, Chemical precipitation can be used to remove heavy metals but the method is not effective enough to meet the discharge effluent standards. For wastewater treatment, the ion exchange process is an efficient method but it is not widely used because of its higher cost. Flocculation or coagulation needs to accompany precipitation, which usually results in huge maintenance costs for handling despite large volumes of sludge containing heavy metal ions. Despite having effectiveness in metal removal, the time consumption of the biological sorption process is high and the regeneration of the bio-sorption material is not possible (Ramakrishna and Viraraghavan, 1997).

In light of the facts, researchers are investigating better and alternative effective methods for the treatment of heavy metal-containing wastewater. As a result of the investigation of the safe and cost-effective method, adsorption has emerged as a better option for treatment of the wastewater. The major benefits of the adsorption technique are lower initial and operating costs for heavy metal removal from wastewater, there is no trouble with the design, and fewer necessities for the control system (Malik, 2004). Compared to the other methods, Adsorption is the most effective and quick method regarding the concentrations of the heavy metal and the volume of the water that it is dissolved in. An increase in the concentration of a particular component at the surface or interface between two phases is known as adsorption (Faust and Aly, 1987). The physical adsorption process does not involve the sharing or transfer of electrons and binding of adsorbate onto the adsorbents caused by van der walls attraction occurs, whereas chemical sorption involves chemical bonding onto the surface of adsorbents (Tripathi and Ranjan, 2015). The substance that is removed from the liquid phase is called the adsorbate and the adsorbent is the solid, liquid, or gas onto which the adsorbate accumulates. There are different types of adsorbents for removing heavy metals effectively from wastewater that are both commercial and bio-adsorbents. The cost of the adsorption process is affected by the cost of the adsorbents. Compared to commercial synthetic adsorbents, the cost of natural adsorbents is much less (Gupta and Babu, 2008). For this reason, importance is given to finding available natural adsorbents for the removal of heavy metals. Removal of heavy metals using waste products is getting popularity because of several sources of material such as organic materials, agricultural waste, industrial by-products, etc. Several studies were done on plant waste such as neem bark (Bhattacharya *et al*., 2006) rice husk (El-Said *et al*., 2012), tea leaves (Ahluwalia and Goyal, 2005), etc. to find their adsorption capacity for removing toxic heavy metal.

Coconut waste can be used as an adsorbent because of its availability in plenty and sorption properties. Its sorption properties are due to the presence of coordinating functional groups such as hydroxyl and carboxyl (Tan *et al*., 1993). The coconut husk is the fibrous layer of the fruit inside the outer shell of a coconut. Coconut husk is processed during the extraction of the long fibers from the shell. It is the binding material that comes from the fiber fraction of the coconut shell (Vishnudav *et al*., 2005). Ash adsorbents produced from coconut fiber husks have higher adsorption capacity due to the high lingo-cellulose material present in them. The maximum 89% removal efficiency of copper using coconut husk is found (Thapak *et al*., 2016). Jin *et al*., 2013 converted coconut into activated carbon and then grafted it with tatraoxalyl ethelenediamine melamine chelate for Cadmium removal along with other heavy metals. Aravind *et al*., 2017 found that coconut coir has good adsorption capacity in removing copper, nickel, and cadmium from industrial wastewater, and removal efficiency is higher at a minor dosage of the adsorbent.

The results of the previous studies showed that coconut fibre ash could be a good adsorbent in wastewater treatment for removing heavy metals. In the current study, an adsorbent prepared from the ash of coconut fiber husk was used to evaluate the efficiency of removing Pb, Cu, and Zn metals from wastewater samples. The objectives of the study were (i) To evaluate the effectiveness of coconut fibre ash as adsorbents in heavy metal removal and (ii) To determine the most effective particle size, dose, and contact time of adsorbents in heavy metal removal. As an unconventional natural adsorbent coconut fibre ash has natural advantages other than being abundant in nature. These materials need less prior processing and are locally available. For this reason, these could be alternatives to other synthetic adsorbents. Following the advantages, this study could be significant to increase the need for wastewater treatment through the use of natural and low-cost adsorbents. And this could play a significant role to prevent the release of heavy metal concentrations into the water bodies, which can cause odor and toxicity, posing potential hazards to human health and the environment.

MATERIALS AND METHODS

Collection of the coconut coir husk

In this study, coconut coir husk was used for the preparation of the adsorbent. As coconut can be found in plenty at our local market, it is easy to collect coir husk or fiber husk at a reasonable price. The coconut fibre husk was collected from an agricultural waste processing institute located in the Mymensingh area, for the preparation of adsorbent.

Cleaning of the material

After the collection of the material, the coconut husk was

Table 1. Initial concentration of the heavy metal Pb, Cu, and Zn in the collected wastewater sample.

Heavy metals	Initial Concentration (mg/L)
Pb	0.545
Cu	9.971
Zn	8.012

cleaned with tap water thoroughly to remove any possible strange particles such as sand, dust, or dirt particles. After cleaning with tap water, the material was washed with distilled water several times. Then the sample was sun-dried for 24 hours. After that, the coconut husk was soaked in a 5% HCl solution for 24 hours (Aravind *et al*., 2017).

Preparation of adsorbent

After completing the soaking process, the coconut husk samples were washed with water again and sun-dried. Then, the husk was taken to a drying oven at 105°C for 24 hours. After drying, the samples were cooled at room temperature. The dried coconut husk was taken into the crucible and placed in a muffle furnace for activation of the carbon. The activation process could be carried out by setting the furnace temperature at different values such as 450°C, 500°C, 550°C, 600°C, etc. (Onyeji *et al*., 2011). This experiment was done by setting the furnace temperature at 450°C. The samples were taken out of the muffle furnace after 20 minutes and cooled in a desiccator. The ash product was then powdered into fine particles. The optimum particle size of coconut husk was 150-200 microns for the removal of heavy metal iron, cadmium, and copper (Paul *et al*., 2017). In this experiment, the particle size of coconut fibre husk or ash was chosen 50 to 200 microns to investigate their adsorption capacity in removing heavy metals Copper (Cu), Zinc (Zn), and lead (Pb). The adsorbent materials of different particle sizes of 50 microns, 100 microns, and 200 microns were separated through sieving operation and then stored in separate plastic bottles for further use.

Wastewater sample collection

The wastewater sample used was collected from the effluent discharge point of the local electroplating industry in the Mymensingh area. It was then carefully contained in the plastic container, which was immediately taken to the laboratory to analyze the initial concentration of Copper (Cu), Zinc (Zn) and lead (Pb) ions (Table 1) in the wastewater sample using Atomic Adsorption Spectrophotometer (AAS) in the Interdisciplinary Institute for Food Security (IIFS) laboratory.

Adsorption study

Adsorption experiments were carried out by adding various amounts of adsorbent (0.2 g, 0.6g, 1g) of different particle sizes (50 microns, 100 microns, 200 microns). The previously prepared adsorbent (activated carbon) was added with 50 ml of the wastewater sample at different dosages. Using a rotary shaker of the Professor Mohammad Hossain Central Laboratory (PMHCL) of the Bangladesh Agricultural University, the adsorbent was mixed with the wastewater sample at 150 rpm at room temperature 29°C. Following different agitation times from the lower 20 min (Aravind *et al*., 2017) to the highest 120 min (Bernard *et al*., 2013) in the previous study, 1 hour agitation time was chosen in this study. Conical flasks were then taken out of the rotary shaker and put aside for 2 hours to allow the adsorbents to adsorb the heavy metal (Aravind *et al*., 2017). The suspension was then filtered using Whatman filter paper and 20 ml of the filtered solution was taken into a test tube as input to the Atomic Adsorption Spectrophotometer (AAS) to find out the final concentration of Cu, Zn, and Pb.

The metal ions adsorbed by the adsorbent were determined using the equation (1) (Chowdhury *et al*., 2011).

$$qt = \frac{(C_o - C_t) \times v}{w} \quad (1)$$

where Co= Initial concentration of the heavy metal in the wastewater sample (mg/L); C_t = Final concentration of the heavy metal after adsorption for time t (mg/L); V = Volume of wastewater used (ml); W = The mass of the adsorbent used (gm)

The percentage of removal efficiency was obtained from the equation (2) (Chowdhury *et al*., 2011).

$$R\ (\%) = \frac{(C_0 - C_t) \times 100}{C_o} \quad (2)$$

Adsorption studies were conducted to examine the effect of adsorbent dosage, particle size, and contact time on the adsorption of Cu, Zn, and Pb from wastewater.

Sample preparation with different adsorbent dosage

The wastewater sample of 50 ml solutions was taken in three different conical flasks at three different adsorbent dosages *i.e.* 0.2 g, 0.6g, and 1g following the previous study (Thapak *et al*., 2016). The Adsorbent of 200 microns particle size was taken in this stage. After the shaking operation at 150 rpm for 1 hour, the flasks were taken out from the rotary shaker and the filtration was done after 2 hours. The filtered content was analyzed for metal concentration.

Sample preparation varying particle size of adsorbent

In this step, adsorption studies were carried out to find the effect of different particle sizes at different adsorbent dosages. Adsorption studies were carried out in three different batches with three different adsorbent dosages to find out the optimum particle sizes at the optimum adsorbent dosage. There were three different conical flasks with 50 ml wastewater samples in every batch. The three flasks of the first batch were under 0.2 g adsorbent dosage with the particle size of 50 microns, 100 microns, and 200 microns respectively. Similarly, with 0.6 g adsorbent dose the three flasks of the second batch and with 1 g adsorbent dose three flasks of the third batch were varied at 50 microns, 100 microns, and 200 microns particle size respectively. 150 rpm shaking speed for 1 hour and contact time for 2 hours were constant in this step. All the samples were filtered using the Whatman filter and filtered samples were taken to AAS for determination of final concentrations.

Preparation of sample varying contact time

To study the effect of contact time on the removal of heavy metal ions three different contact times 40 minutes, 80 minutes, and 120 minutes were chosen for adsorption studies with different adsorbent doses (Bernard *et al.*, 2013). The whole study was done in three different batches where there were three different conical flasks with the 50 ml wastewater sample in every batch. To find out the optimum contact time at optimum adsorbent dosages, the water of the three flasks of every batch was mixed with three different amounts of adsorbent dosages. The solution of the three flasks of the first batch was mixed with 0.2 g adsorbent of 200 microns particle size at 150 rpm for 1 hour. The solution of the first flask was filtered after 40 minutes, the second one was done after 80 minutes and the third one was done after 120 minutes. Similarly, adsorption was done with a 0.6 g adsorbent dose in the three flasks of the second batch, and in the three flasks of the third batch, adsorption was done with a 1 g adsorbent dose. After the shaking operation three different contact times 40 minutes, 80 minutes, and 120 minutes were chosen for the second and third batches following the same procedure of the first batch. Finally, all the filtered sample was taken to AAS for determination of the metal ion concentration.

RESULTS AND DISCUSSION

Effect of adsorbent dosage

The adsorbent dosage was varied from 0.2 to 1 g at specific parameters (200-micron particle size, 150 rpm shaking speed for 1 hour, contact time 2 hours). Figure 1 shows that the percent removal efficiency increases with increasing adsorbent dosages. Pb ion attains maximum removal efficiency at 1gm dosage with 95.04% removal efficiency. With increasing adsorbent dosage, the increased removal efficiency was attained for Cu and Zn ions also. The highest 80% and 34% removal efficiency was attained at 1g adsorbent dosage for Cu and Zn, respectively as shown in figure 1. It was also reported that there was a very slight increase in removal efficiency from 0.6g to 1g adsorbent dosage and almost maximum removal efficiency was attained before reaching a maximum adsorbent dosage of 1g. The maximum removal efficiency of 88% was found for Cu removal with 1g adsorbent of coconut husk (Thapak *et al.*, 2016). Aravind *et al.*, 2017 reported that a minor dosage of 1g adsorbents from coconut coir was effective for Cu, Ni, and Cd removal and there were very minor changes in removal with increasing adsorbent dosage up to 5g. Onyeji *et al.*, 2011 determined the adsorption capacity of adsorbents prepared from coconut shells. In their study, 80% removal efficiency was found in removing Hg(II) and Pb(II) at low adsorbent dosage of 2 g, and 29% removal efficiency was found in removing Cu(II) metal ions at an adsorbent dosage of 2 gm.

Effect of particle size

To find the effect of different particle sizes at different adsorbent dosages, adsorbents of particle sizes ranging from 50 to 200 microns were used at this stage.

Effect of particle size (at 0.2 g adsorbent dosage)

The percent removal efficiency at a different particle size of the adsorbent with a specific 0.2 g adsorbent dosage is shown in the following Figure 2. The fig shows that the removal efficiency increases with increased particle size. The lowest removal efficiency of 10% was attained at the lowest particle size for Zn metal ions. Maximum removal of Cu and Zn ions was attained at particle size ranges of 200 microns with 70% and 88%, respectively. It was reported that satisfactory results were not found at 50-micron particle size for Pb, Cu, and Zn metal removal. The maximum removal efficiency was attained at 200-micron particle size. It was explained by Eze *et al.*, 2013, that sorption is controlled by particle diffusion and the particle size varies with the number of micropores in their surface area, and, the increase in micropores increases the number of accessible sites which increases the amount adsorbed.

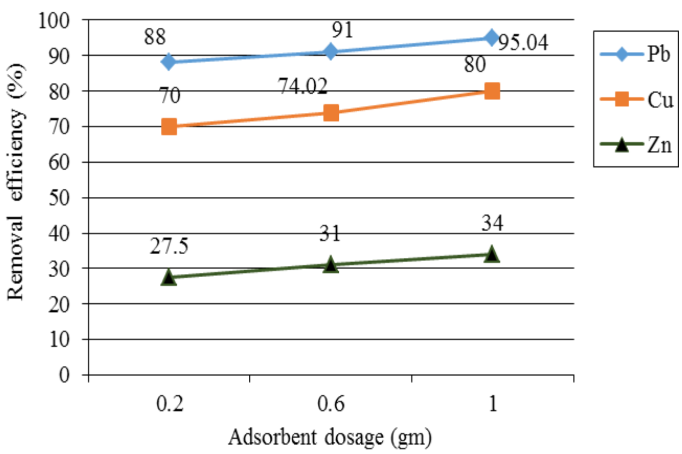

Figure 1. *Effect of adsorbent dosage on adsorption of heavy metals Pb, Cu, and Zn.*

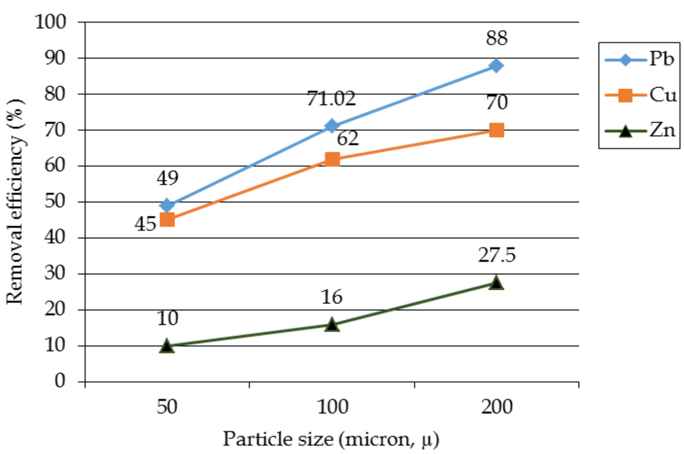

Figure 2. *Effect of particle size (at 0.2g adsorbent dosage) on adsorption of heavy metals Pb, Cu, and Zn.*

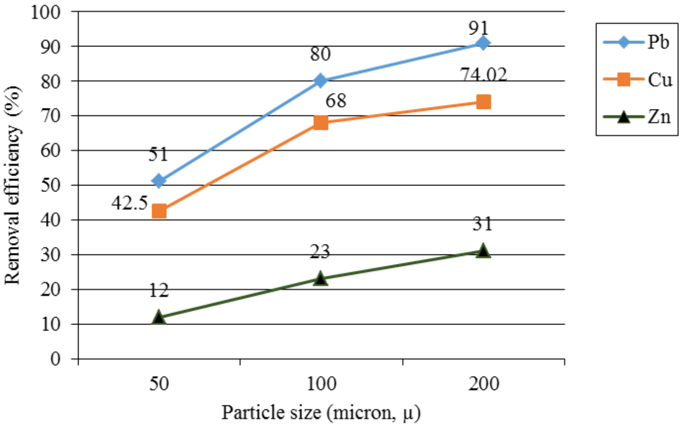

Figure 3. *Effect of particle size (at 0.6g adsorbent dosage) on adsorption of heavy metals Pb, Cu, and Zn.*

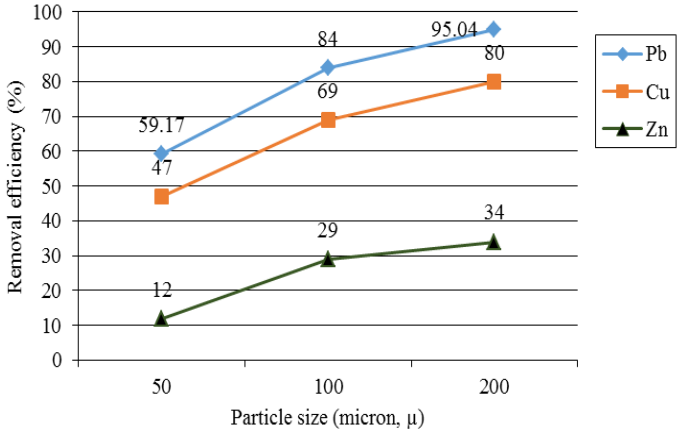

Figure 4. *Effect of particle size (at 1g adsorbent dosage) on adsorption of heavy metals Pb, Cu, and Zn.*

Effect of particle size (at 0.6 g adsorbent dosage)

The percent removal efficiency at a different particle size of the adsorbent with a specific 0.6 g adsorbent dosage is shown in the following Figure 3. and shows that the removal efficiency increases with increased particle size. For Zn ion, an adsorbent with the lowest particle size of 50 microns results in lower adsorption of metal ions with only 12% removal efficiency. A good adsorption capacity at particle size ranges 100 and 200 microns is indicated for Pb ion with 80% and 91%, respectively at a specific 0.6 gm dosage. For Pb and Cu, removal efficiency increases with increasing particle size and also adsorbent dosage. A Similar trend was observed in the previous study that the optimum particle size of coconut husk adsorbent was 150-200 microns for Fe, Cu, and Cd metal removal (Paul *et al*., 2017). Here at 0.6g adsorbent dosage, the maximum removal efficiency of 74.02% is found at a maximum particle size of 200 microns for Cu ion. So, adsorbents prepared in this study from coconut fibre ash gave satisfactory results from 100-200 microns particle size. On the other hand, the suitable particle size of adsorbents prepared from mango peel was 400-500 microns for Fe, Cu, and Cd metal removal (Paul *et al*., 2017).

Effect of particle size (at 1 g adsorbent dosage)

At a specific 1 gm adsorbent dosage, the percent removal efficiency increases with increasing particle size. Figure 4 shows that the higher removal efficiency could not be attained at a lower particle size with an increased 1gm adsorbent dosage. Again similar lowest removal efficiency of 12% is obtained for Zn ion at 50 microns particle size with an increased 1gm adsorbent dosage compared to the previous adsorbent dosage (0.6gm) for the same particle size. Similarly, maximum 95.04% and 80% removal efficiency is obtained at a maximum particle size of 200 microns for Pb and Cu ions, respectively. It was reported that an optimum particle size of 200 microns showed maximum removal efficiency at 1gm adsorbent dosage which indicated that to get the best results from coconut fibre ash adsorbents 1 gm adsorbent dosage of 200 microns particle size should be used.

Effect of contact time

The effect of contact time on the removal of heavy metal ions was studied under three different contact times 40 minutes, 80 minutes, and 120 minutes.

Effect of contact time (at 0.2 g adsorbent dosage)

The effect of different contact times such as 40 minutes, 80 minutes, and 120 minutes, on the removal of metal ions was studied under certain conditions (0.2 gm adsorbent dosage, 200 microns particle size, 150 rpm of shaking speed for 1 hour). Figure 5 shows that with increasing contact time, the percent removal efficiency increases. It also shows that different heavy metals attained maximum removal efficiency at different contact times. For Pb ions, the maximum removal efficiency of 88% is obtained at 80 minutes of contact time and there was no increase in removal ions with further increase in contact time. Sometimes no significant changes were identified in the percentage removal with further increase in time that was observed. Observing a similar trend Cherono *et al*., 2021 reported that the adsorption sites were saturated due to intense competition among the heavy metals studied. The maximum removal efficiency of 70% and 27.5% is obtained at a maximum 120 minutes contact time for Cu and Zn ions respectively which could not be attained at lower 40 minutes contact time.

Effect of contact time (at 0.6 g adsorbent dosage)

The relationship between percent removal of heavy metal and contact time at a specific 0.6 g adsorbent dosage is shown in the following Figure 6. where the percent removal efficiency increases with increasing contact time at 0.6 g adsorbent dosage. There is a slight increase in Cu ion removal from 77.4% to 79% with increasing contact time. Thapak *et al*., 2016 also reported nearly similar results 81% throughout the 120 minutes contact times for Cu removal using 0.6 g adsorbents prepared from coconut husk. 90.09% removal efficiency occurs at 40-minute contact time for Pb ion, from where further increase in time results in desorption with decreasing 88.91% efficiency and again maximum 93% efficiency is attained at 120 minute contact time. It happened due to the desorption from the adsorbent surface. Due to the availability of the uncovered surface area of the adsorbents, adsorption first increases with increasing contact time and the reactive sites are progressively filled where the adsorption of heavy metal ions takes place which creates

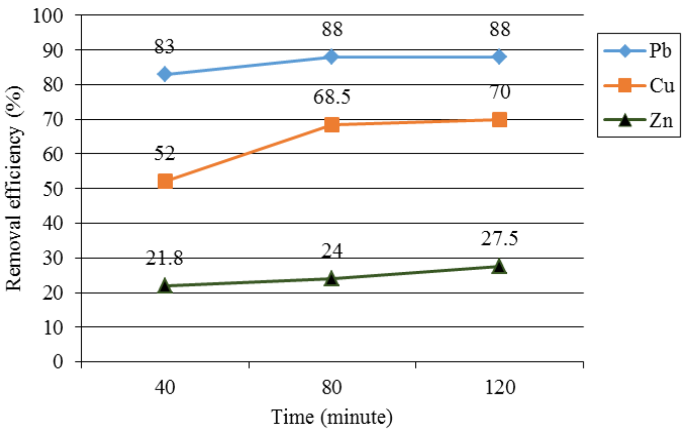

Figure 5. *Effect of contact time (at 0.2 g adsorbent dosage) on adsorption of heavy metals Pb, Cu, and Zn.*

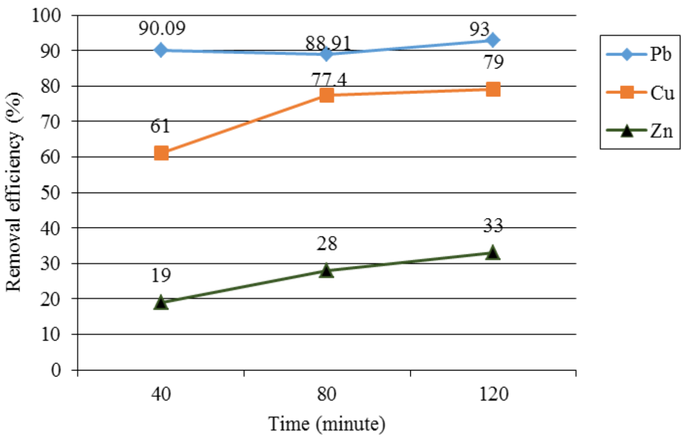

Figure 6. *Effect of contact time (at 0.6 g adsorbent dosage) on adsorption of heavy metals Pb, Cu, and Zn.*

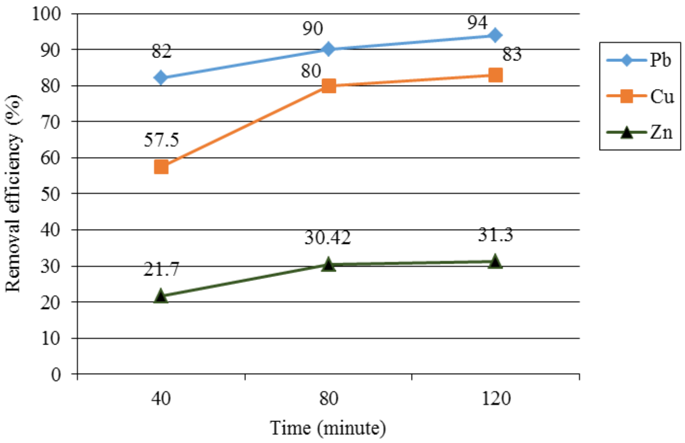

Figure 7. *Effect of contact time (at 1g adsorbent dosage) on adsorption of heavy metals Pb, Cu, and Zn.*

difficulty in sorption and results in desorption (Thajeel *et al.*, 2013). It was identified that to ensure maximum adsorption of heavy metals, adsorption should be allowed on a maximum contact time of 120 min. Maximum removal efficiency is also obtained for Zn ion with increasing contact time.

Effect of contact time (at 1 g adsorbent dosage)

At 1gm adsorbent dosage, the maximum removal efficiency is obtained at a maximum contact time of 120 minutes. Figure 7 shows that the maximum 94% and 31.3% removal efficiency are obtained at an increased 1g adsorbent dosage for Pb and Zn ions. For Zn metal, significant changes were not identified increasing both contact time and adsorbent dosage. But, the lowest removal efficiency 57.5% at 40 minutes increases to the highest at 83% at 120 minute contact time for Cu ion. Increased contact time of 120 minutes with 1g adsorbent dosage facilitates the maximum removal efficiency of Pb and Cu. The maximum 120 minutes contact time was also considered in the adsorption study of Cr, Cd, and Pb ions using *E. camaldulensis* activated carbon where it was reported that the higher the contact time between the sorbent surface and the metal solutions, the more complete adsorption by activating the equilibrium (Gebretsadik *et al.*, 2020).

Conclusion

The study focused on the laboratory evaluation of the performance of the coconut fibre ash in the removal of Pb, Zn, and Cu ions from wastewater. The adsorption capacity of the adsorbents provided satisfactory performance in removing heavy metals. The maximum removal efficiency of 95.04%, 80%, and 34% was obtained at the maximum amount (1 gm) of adsorbent dosage for Pb, Cu, and Zn, respectively. So, it could be concluded that a 1g adsorbent dosage of coconut fibre ash will give satisfactory results for Pb and Cu metal removal. The Optimum particle size for maximum removal efficiency was identified from 100-200 microns at 1 gm dosage which reached the highest value for 200 microns particle size. So, 200 microns particle size of adsorbents from coconut fibre ash would be suitable for Pb, Cu, and Zn removal from wastewater. In the case of the contact time, the maximum removal efficiency is found at a 1 g adsorbent dosage with a contact time of 120 minutes. To ensure the highest removal adsorption should be allowed with a 1g dosage at 120 minutes contact time. In most of the cases, higher efficiency was obtained at 80 minutes and for further increases in time very small changes occur. Sometimes desorption from the surface of the adsorbents was identified which indicated that to ensure maximum removal of heavy metal, adsorption should be allowed on maximum contact time of 120 minutes. Considering the desorption issues, to ensure maximum removal efficiency the highest 120 minutes of contact time would be allowed during the adsorption process with coconut fibre ash adsorbents. Further study could be recommended by varying other parameters such as shaking speed, pH, and temperature.

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Conflict of interest

The authors declare there is no conflict of interest regarding the publication of this paper.

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