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Regular Research Manuscript

Use of activated palm kernel shells on adsorption of heavy metals (Cu^{2+} and Pb^{2+}) from contaminated wastewater

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ABSTRACT

This research aimed to evaluate the adsorption capacity of low-cost agricultural waste adsorbent-activated palm kernel shells (APKS) for the removal of Pb^{2+} and Cu^{2+} ions from synthetic wastewater that had the concentration of copper and lead ions comparable to that of waste produced in Tanzanian research and teaching laboratories. This study was carried out in batch and continuous column adsorption reactors. The study examined the influence of particle size (0.71 mm–2.36 mm), pH (4.5–8.6), adsorbent dose (5–20 mg/L) and initial concentration C_i (1.93– 4 mg/l) on APKS adsorption removal efficiency. The batch results indicated that under alkali conditions (pH 8.4 -8.6), APKS has an adsorption removal efficiency of 77.3% - 92% and 98.2-98.5% for Cu^{2+} and Pb^{2+} ions, respectively. Under acidic conditions (pH: 4.0 - 4.5), the APKS has a lower adsorption capacity of 23.7% and 36.29% for Cu^{2+} and Pb^{2+} ions, respectively. The column test results showed that APKS had a removal efficiency of 98.4% and 73.6 for Pb^{2+} and Cu^{2+} ions, respectively. The results also showed that both Langmuir and Freundlich adsorption isotherms were the best model for the adsorption of Cu2+ ions on APKS, with R^2 of 0.997 and 0.97, respectively. The results also showed that APKS can be utilised to treat wastewater with Pb^{2+} and Cu^{2+} concentrations to meet the effluent discharge standards. The study recommends using activated palm kernel shells to remove heavy metals in wastewater before discharging them into the environment.

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INTRODUCTION

Wastewater containing heavy metals must be treated to meet discharge standards before being discharged into the existing wastewater treatment systems or receiving water bodies. When wastewater with high concentrations of heavy metals is discharged into the existing sanitation system, it affects the performance of microorganisms in the biological treatment systems. It thus reduces the efficiency of the biological treatment process (Tekere *et al*., 2016). According to the literature, waste generated from institutions and research contains heavy metals. They are currently discharged into existing domestic wastewater systems without proper treatment, thus posing a high risk to people and the environment (Kihampa, 2015). Unlike organic pollutants, whose majority are susceptible to biological degradation, heavy metal ions do not degrade into harmless end products and thus have

created significant environmental issues (Hegazi, 2013).

This practice of discharging untreated wastewater above the allowable discharge limit contradicts target 6.3 of the Sustainable Development Goals to reduce pollution, eliminate dumping and minimise release of hazardous chemicals and materials. In particular, the presence of heavy metal ions is a significant concern due to their toxicity to many life forms. When heavy metals continuously accumulate in the human body, it can lead to severe adverse effects such as brain damage, skin diseases, liver damage, kidney failure, anaemia, hepatitis, ulcers and carcinogens (Vardhan *et al*., 2019). In addition, when heavy metals accumulate in the soil, it hinders plant growth, and when above the allowable discharge limits, it may also affect aquatic organisms in receiving water bodies.

Furthermore, the literature shows that copper (Cu^{2+}), zinc (Zn^{2+}), lead (Pb^{2+}) and cadmium (Cd^{2+}) are the elements of most environmental concern and are commonly reported to contaminate soil and water and food chain (Musharafi *et al*. 2013). A study conducted in research and educational laboratories indicates that heavy metal concentrations in wastewater can reach 3.44 mg/L and 3.22 mg/L for copper and lead, respectively, higher than allowable discharge limits. Therefore, to circumvent the problem of heavy metals, it is essential to treat wastewater to meet the discharge standards and protect the environment.

Several methods exist to remove heavy metals from wastewater, including ion exchange, chemical precipitation, membrane filtration, solvent extraction, and electrochemical treatment (Shaikh *et al*., 2018). However, these techniques have limitations of high operation costs and secondary pollution due to the addition of chemicals in treatment and low efficiency achieved (Ding *et al*., 2016). Among several treatment options available, studies recommend applying an adsorption process with activated carbonaceous materials such as coconut shells, palm kernel shells, chickpea husks, corn cobs, banana leaves, and rice husks to remove heavy metals in water and wastewater (Samuel *et al*., 2016).

Previous studies have indicated that the adsorption process using activated carbon is the most efficient and cost-effective technique (Ali, 2012). Furthermore, activated carbon prepared from agriculture is a suitable adsorbent for most pollutants (Foo and Hameed, 2012). Despite past studies on activated carbon to remove heavy metals, studies on palm kernel shells are particularly limited. This study aimed to use environmentally friendly and economically advantageous physically activated carbon from palm kernel shells (APKS) as adsorbent materials to treat (Cu^{2+}) and (Pb^{2+}) ions from wastewater. The study used synthetic solutions containing copper and lead ions concentrations comparable to that of waste produced in Tanzania's research and educational laboratory. The selection of palm kernel shells was based on the fact that they are cheap and readily available as by-products of palm oil plants. Although there is still little research on using palm kernel shells, according to (Rabia *et al*., 2018), palm kernel shells (PKS) are effective in adsorbing Cr^{6+}, Pb^{2+}, Cd^{2+} and Zn^{2+} ions.

MATERIALS AND METHODS

Preparation of the Synthetic Wastewater

The chemicals used to prepare synthetic wastewater of cu and pb were obtained from the University of Dar es Salaam water laboratory. Synthetic wastewater with Cu^{2+}ions was prepared using hydrated copper sulphate ($CuSO_4.5H_2O$). A stock solution of concentrated copper sulphates ($CuSO_4$) was prepared by dissolving 5.89 g of hydrated copper sulphate into 0.5 litres of distilled water to make a concentrated solution of 3000 ppm solution, and its concentration was tested in a laboratory using the Palin test photometry method. Synthetic wastewater with (Pb^{2+}) was prepared using Lead nitrate ($PbNO_3)_2$, where 5.6 mg was required to make a 3.5 mg/L concentration. To prepare the Cu^{2+} concentration of 400, 3800, 2600,

and 2650 ppm, the stock solution of copper sulphate and distilled water was mixed (volume by volume) for 15 minutes at the ratios of 5.7:5000, 5.4:5000, 3.8:5000, 3.7:5000: respectively. The Palin test using a 7500-interface photometer (Gateshead NE ONS, UK) was used to measure the copper concentration of the prepared synthetic wastewater. The prepared synthetic wastewater, each 5 litres, was then stored in a conical flask before being treated by an adsorbent.

Similarly, the Pb^{2+} concentrations of 3500, 3415, 2130 and 1930 ppm were prepared and stored before the experiment. The Pb^{2+} concentrations were measured using Perkin Elmer Analyst 100 Atomic Absorption Spectrophotometer at Ardhi University Laboratory, Tanzania. For synthetic wastewater with mixed Cu^{2+} and Pb^{2+} using a dilution factor established, a 20 L solution was prepared by dissolving 0.112 g of Pb $(NO_3)_2$ and 24 ml of $CuSO_4$ concentration was added into the same 20-litre Plastic container, followed by mixing to obtain a concentration of 2.13 mg/l Pb^{2+} and 2650 ppm Cu^{2+} solution which was used for treatment testing for solution contain both heavy metals. The required volume was taken from the big flask into a testing flask with a capacity of 250 ml for batch testing using a prepared activated palm kernel shell as an adsorbent.

Collection and Preparation of Adsorbents

Precursor materials collection

Precursor materials of palm kernel shells from the plant (palm oil process by-product) of about 65 kg were collected from Tans Soap Enterprises Limited in the Kigoma region (4° 52'43.5' 'S 29°38'59.2"E). Following the collection into sacks, the palm kernel shell materials were transported to a water resources laboratory, where they were washed with tap water to remove ditty and oils from the palm shells. Then, palm kennel shells were dried by the sun for five consecutive days before being processed into activated carbon.

Preparation of the activated carbon adsorbents

The carbonised reactor at the University of Dare Salaam (Steam activation reactor) (Currently stationed at Goba) was used for the carbonisation process using a physical activation process, which includes mainly carbonisation at about 600°C followed by steam activation processes. During the activation process, the carbonised palm kernel shell materials were first crushed into small sizes using a crushing machine available at the plant unit and then into small particles up to 2.5 mm. Then, the crushed carbonised palm kernel shell was sieved with a sieve of 2 mm at the plant to remove dust materials. The sieved materials were placed into an activation reactor and were activated for about 5-6 hours with steam at high temperatures. Then, the activated palm kernel shell was allowed to cool for 12 hrs, which was overnight, where they were then overloaded from the reactor the following day. After the activation process, the particle size was measured using sieve analysis at the structural laboratory of the University of Dar es Salaam. The APKS were sieved to get different surface diameters 0.71 mm, 1 mm, 1.4 mm, 1.7 mm, 2 mm, and 2.3 mm, packed into 2 kg plastic bags and stored at 4 ℃ at the water laboratory before being used as adsorbent for adsorption. Ash contents and moisture contents of APKS were measured gravimetric according to the standard methods for examinations of water and wastewater samples (APHA 2012). About 65g of APKS were placed into a 100 ml calibrated beaker to determine apparent density, and the change in volume in the beaker was measured at 150 ml. The apparent density was determined as the ratio of the mass of activated palm kernel shells to the volume of the displaced water (Equation 1) (Verla *et al*., 2012).

Apparent density = Mass of APK (*g*)/Displaced Water Volume (ml) (1)

Batch adsorption experiment

A batch adsorption experiment was carried out in the laboratory to determine the

capacity of activated palm kernel shells to remove Cu^{2+} and Pb^{2+} ions from the wastewater. The experiment was carried out in a 250 ml conical flask using 150 ml of wastewater with Pb^{2+} and Cu^{2+} initial concentrations of 1.93, 2.13, 3.415 and 2.6, 3.5, 3.8 and 4 mg/150 ml, respectively. The mixture of adsorbent and adsorbate was agitated at 200 rpm using the IKA KS 4000 shaking machine to facilitate adsorption. The study assessed the influence of Contact time of 20, 40, 60 and 80 minutes, adsorbent dosage of 5, 10, 15 and 20 mg/150ml and pH 4, 4.5, 8.4 and 8.6. The pH adjustments were carried out using HCL and KOH Moles solutions. After each of the contact time tests, the sample was filtered using Whatman filter paper 110 mm diameter, and the filtrate obtained was tested to know its concentration at each contact time using the Palin test method by 7500 interface photometer (Gateshead NE ONS, UK) for Cu^{2+}ion concentrations. On the other hand, the Perkin Elmer A Analyst 100 Atomic Absorption Spectrophotometer was used for testing Pb^{2+}ion concentrations.

Treatment Efficiency Analysis

The percentage treatment removal efficiency R_e (%) for the experimental results was determined as the percentage ratios of the difference between initial metal concentration and concentration after the adsorption (C_i - C_e) to the initial metal concentrations (equation 2). On the other hand, the amount of metal adsorbed per unit mass at equilibrium was calculated using the relation as indicated in equation 2.3 (Desta 2013).

$$R_e\ (\%) = (C_i - C_e/C_i)x100 \qquad (2)$$

$$Qe = (C_i - C_e)V/M \qquad (3)$$

where R_e (%) = Removal percentage; *Ci* = Initial concentration (mg/*L*); C_e = Concentration after adsorption (mg/*L*); *Qe* = metal adsorbed per unit mass at equilibrium; *V* = The volume of the solution (ml); M = mass of the adsorbent (*g*)

Adsorption Isotherm

This study used the Langmuir and Freundlich isotherm models to describe the metal ion distribution between the liquid and solid phases. The Langmuir isotherm model assumes the adsorbate is adsorbed in a monolayer with a fixed number of adsorption sites that have no interaction of the adsorbate molecules in the plane of adsorbent surfaces (Odebunmi 2010). Thus, the saturation point is reached when all adsorption sites are occupied. The Langmuir model is presented in equation 4.

$$q_e = q_{max}(K_L * C_e)/(1 + K_L * C_e) \qquad (4)$$

where q_e = is the quantity of adsorbate adsorbed at equilibrium (mg/L); q_{max} =Maximum possible concentration of adsorbate adsorbed and occupied total adsorbent surface;K_L =Langmuir equilibrium constant; Ce = quantity of adsorbate remaining in solution at equilibrium (mg g^{-1}). The maximum concentration adsorbed and occupied on the total adsorbent surface was calculated using the relation that,

$$q_{max} = \frac{1}{intercept} \qquad (5)$$

The Freundlich adsorption isotherm assumes multilayer adsorption and non-uniform surfaces. The Freundlich constants can be determined by plotting log (x/m) and log C_e, as indicated in equation 6.

$$Log\ \frac{x}{m} = log\ k_f + \frac{1}{n} Log\ C_e \qquad (6)$$

where *x/m* = is the quantity of adsorbate adsorbed at equilibrium per mass of adsorbent (mg adsorbed/gram activated carbon); K_f = Freundlich constant factor (mg absorbate/gram activated carbon). The n values represent nonlinearity in the adsorption process. If $n > 1$, adsorption is a physical process; if $n < 1$, the adsorption is a chemical process; and if n = 1, the adsorption is linear; C_e = quantity of adsorbate remaining in solution at equilibrium (mg l^{-1}) (Ayawei *et al*., 2017).

Column experiment

The column adsorption experiments were carried out in a column of activated palm kernel shells, five layers arranged with surface area increasing from 1 mm, 1.4 mm, 1.7 mm, 2 mm, and 2.36 mm to the top. The synthetic solutions containing 2.65 mg/l Cu concentration and Pb^{2+} concentrations of 3.41 and 2.13 mg/l were used in the experiment. The synthetic water was treated upward through the adsorption column, and the treated samples were taken in different layers, 1.4 mm, 2 mm, and a final 2.36 mm layer, for analysis. The collected treated wastewater was analysed for the copper and lead concentration per the method described in section 2.1 above. The experiment was operated for about 5 days at a *pH* of 8.6 and a constant flow rate of 870 *mL*/min.

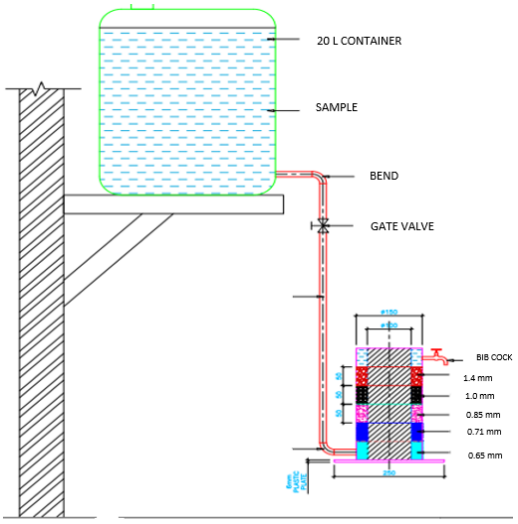

Figure 1: Up flow column experiment set up.

RESULTS AND DISCUSSIONS

Batch Experiment

Effects of adsorption dosage on the adsorption efficiency of Cu $^{2+}$ and Pb^{2+}

It can be seen from Figure 2 that the Cu^{2+} concentrations were lowered to less than 1 mg/l at the adsorbent dosages of 5 g, 10 g, 15 g, and 20 g administered during the experiment. A dosage of 5 g/150 ml resulted in a higher Cu^{2+} concentration adsorption capacity with a 92% removal efficiency at the contact time of 40 minutes. The removal efficiencies of Cu^{2+} concentration for 10 g/150 ml, 15 g/150 ml, and 20 g/150 ml at the contact time of 40 minutes were 83%, 79%, and 66%. The higher Cu^{2+} concentration removal efficiency in this result was attained at a low adsorbent dosage (5 g), and an additional increase in dosage did not affect the adsorbent's ability to increase its adsorption capacity. This could be due to more active sites during the first 20 min of the process, but the adsorption rate slowed as the active sites were filled. After a contact time of 80 minutes, the removal efficiencies for 5 g/150 ml, 10 g/150 ml, 15 g/150 ml, and 20 g/150 ml, respectively, were 91%, 85%, 83%, and 77%. The results presented in Figure 3 demonstrate that, within 20 minutes, the Pb^{2+} concentrations were lowered to 0.072 mg/l, 0.085 mg/l, 0.047 mg/l, and 0.073 mg/l at the adsorbent dosages of 5 g, 10 g, 15 g, and 20 g used in the experiment. A dosage of 15 g/150 ml resulted in a higher Pb^{2+} concentration adsorption capacity with a 98.6% removal efficiency at the contact time of 20 minutes. The respective removal efficiencies of Pb^{2+} concentration for 5 g/150ml, 10 g/150ml, and 20 g/150ml at the contact time of 20 minutes were 97.9%, 97.5%, and 97.9%. Following the eighty-minute contact period, the Pb^{2+} concentrations for 5 g, 10 g, 15 g, and 20 g were 0.061 g/l, 0.055 mg/l, 0.051 mg/l, and 0.05 mg/l. The results of this study show that APKS is more efficient than other adsorbents, such as husk, which had a Pb^{2+} concentration removal efficiency of 22% at an adsorbent dosage of 20g (Hegazi, 2013).

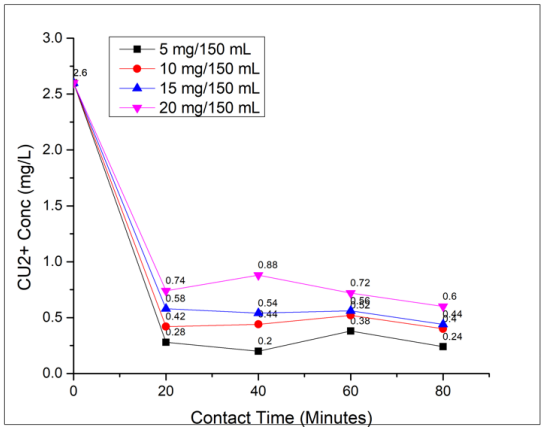

Figure 2: Effects of adsorbent dosage on adsorption of Cu^{2+}.

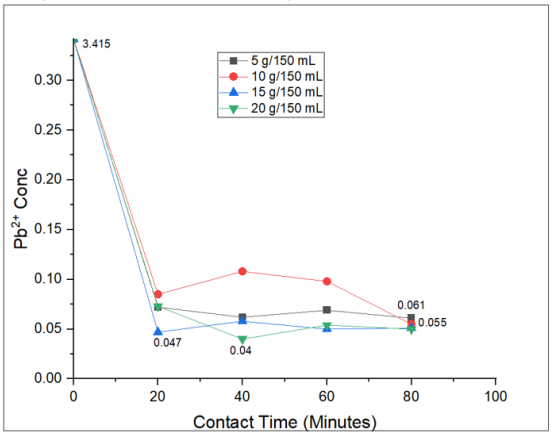

Figure 3: Effects of adsorbent dosage on adsorption of Pb^{2+}.

Effects of initial concentration (C_i) on the adsorption efficiency of Cu^{2+} and Pb^{2+}.

Figure 4 indicates that during the initial 20 minutes of the experiment, the experimental flask, which contained 3.5 and 4 mg/L adsorbent dosages, demonstrated a Cu^{2+} concentration removal efficiency of 77.71% and 78.57%, respectively. On the other hand, the experimental flask containing 3.8 mg/L of adsorbent dosages had the lowest removal efficiency of 58.57% during the first 20 minutes of the experiment. The Cu^{2+} concentration adsorption in all the experimental flasks gradually increased after 20 minutes of contact time. The adsorbent dosages of 2.6 mg/L, 3.5 mg/L, 3. 8 mg/L, and 4.0 mg/L resulted in Cu^{2+} concentration removal efficiencies of 66.21 %, 75.43%, 80.86%, and 84.29% after 80 minutes of contact time, respectively. It can be generalised that the removal efficiencies of copper concentration increased as the amount of adsorbent dosage increased.

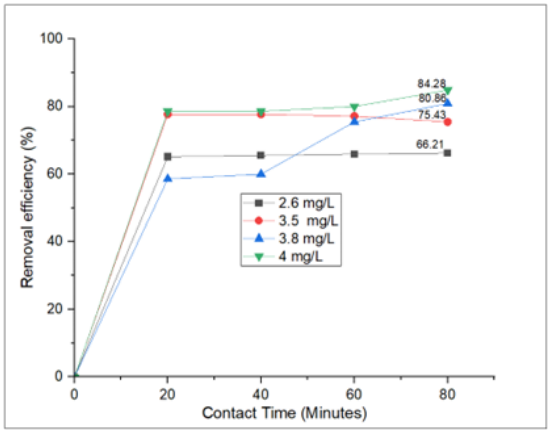

Figure 4: Effects of initial concentration of Cu^{2+}.

In light of Fig. 5, the experimental flask containing 1.93 mg/l, 2.13 mg/l, and 3.15 mg/l adsorbent dosages had a Pb^{2+} concentration removal efficiency of 99.9%, 95.5%, and 95.7%, respectively, during the first 20 minutes of the experiment. The Pb^{2+} concentration adsorption was high during the first 20 minutes because more active sites were saturated by the adsorbate over time, resulting in less ion adsorbed at the higher contact time. Consequently, it can be concluded that the ideal contact time for treating Cu^{2+} and Pb^{2+} concentrations with activated palm kennel shells is 20 minutes.

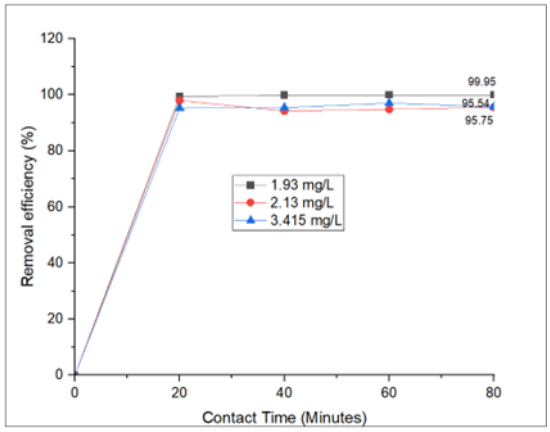

Figure 5: Effects of initial concentration of Pb^{2+}.

Effects of particle size on Pb^{2+} and Cu^{2+} ions adsorption

As can be seen in Figure 6, the Pb^{2+} concentration of 3.415 mg/l was reduced to 0.035 mg/l (about 99% removal) at the first 20 minutes of contact time when using the particle size of 2.00 mm. The respective removal efficiencies of Pb^{2+} concentration for the particle size of 2.36 mm, 1.4 mm, and 0.71mm at the contact time of 20 minutes were 97%, 89%, and 87%. It is clear from the figure that to achieve the required allowable Pb^{2+} discharge standard of 0.1 mg/L, particle sizes of 2.0 mm and 2.36 mm and a contact time of 20 minutes would be necessary. When comparing Figure 6 and 7, the results show that APKS were more successful in adsorbing Pb^{2+} concentrations than Cu^{2+} concentrations. While the Pb^{2+} concentration was reduced from 3.415 mg/l to 0.035 mg/l (approx. 99% removal efficiency), the copper concentration was decreased from 3.8 mg/l to 0.52 mg/l, giving a removal efficiency of approx. 86.3%. With a contact time of 20 minutes, the lowest removal efficiency was achieved for Pb^{2+} and Cu^{2+} at a particle size of 0.71 mm. The fewer active sites available for the adsorption process could be the reason for the lower removal efficiency of the 0.71 mm particle size.

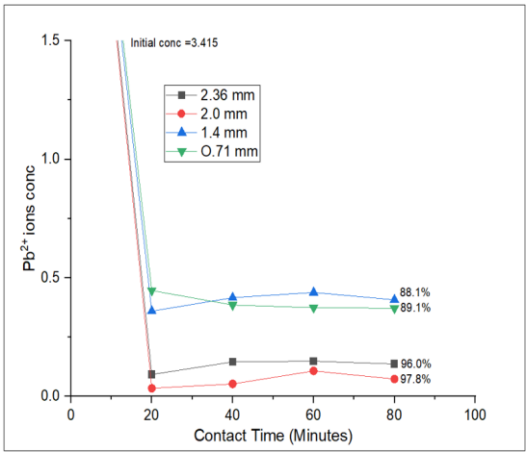

Figure 6: Effects of particle size on removal of Pb^{2+}.

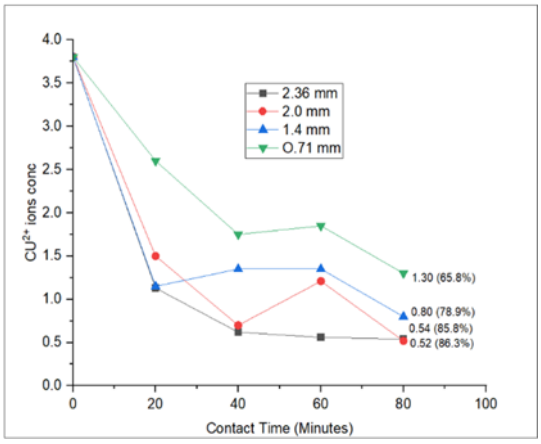

Figure 7: Effects of particle size on removal of Cu^{2+}.

pH effects on adsorption efficiency of Cu^{2+}

Figure 8 shows that APKS were more effective at basic conditions (pH:8.35-8.58) than when operated under acidic conditions (pH:4-4.5). Basic conditions reduced the Cu^{2+} concentration from 2.6 mg/L to 2 mg/L (about 90.7% removal efficiency). The Cu^{2+} concentration, on the other hand, decreased under acidic conditions from 2.6 mg/L to 2 mg/L, which resulted in a 23% removal efficiency. These results can be described by the fact that at lower pH values, the adsorption sites are saturated by H^{+}, lowering the adsorption process. When pH values increase, the adsorption sites become available, increasing the adsorption process (Abbar et al. 2017). Yi et al. (2014) found that the commercial APKS had the maximum adsorption capacity of uranium from aqueous solutions at acidic conditions (pH:5.5). The variations in results can be attributed to the chemistry that different metals have with the functional groups of activated carbon. Thus, the experimental conditions (Basic or acidic) that provided this study's best adsorption results cannot be generalised.

Column test wastewater with Cu^{2+} and Pb^{2+} ions concentrations

As observed in Figure 9, when treated using the continuous column model, APKS were more successful at adsorbing

Pb^{2+}concentration than Cu^{2+}concentration. Similar results were obtained with the batch experiments. The Pb^{2+} concentration was reduced from 2.213 mg/L to 0.093 mg/L, which resulted in 98.4% removal efficiency. On the other hand, the Cu^{2+} concentration was decreased from 2.65 mg/L to 0.70 mg/L, which resulted in a removal efficiency of about 73.6%.

Adsorption isotherms results

An APKS adsorbent mass of 15 g, an adsorbate volume of 150 mL, and a contact time of 80 minutes with the different initial concentrations of copper and lead ions were used to determine the adsorption isotherm. To obtain different equilibrium concentrations for Langmuir, the graph of 1/qe Vs 1/Ce was plotted as shown in Figure 10 and 11 On the other hand, to obtain the Freundlich equilibrium concentrations, the log qe vs. log Ce graph was plotted as shown in Figure 12 and 13.

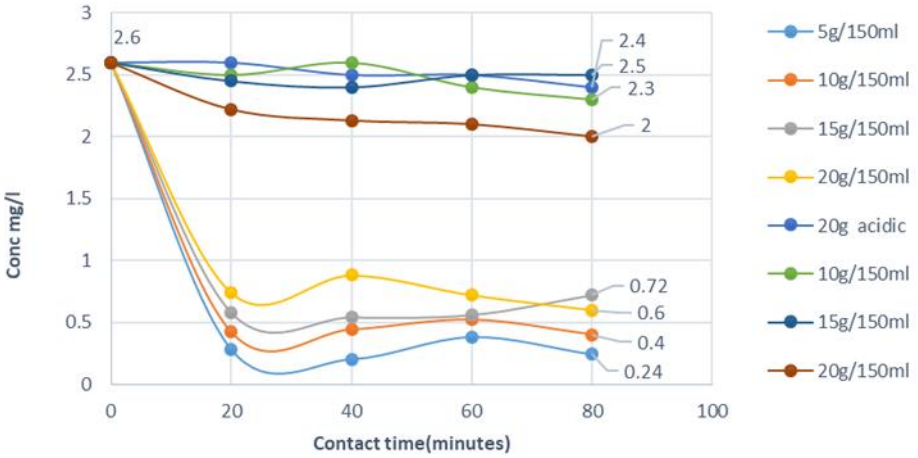

Figure 8: Effects of pH on Adsorption Cu^{2+} ions from wastewater.

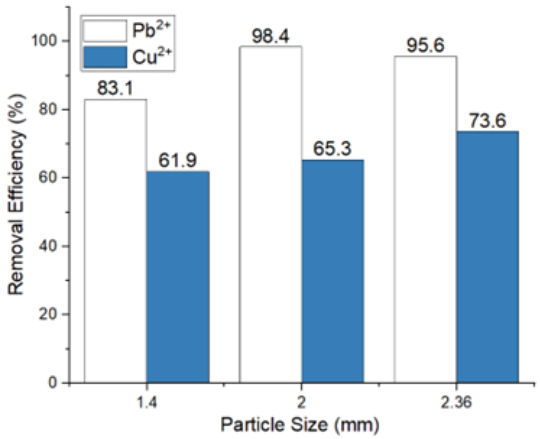

Figure 9: Pb^{2+} and C^{2+}Column tests.

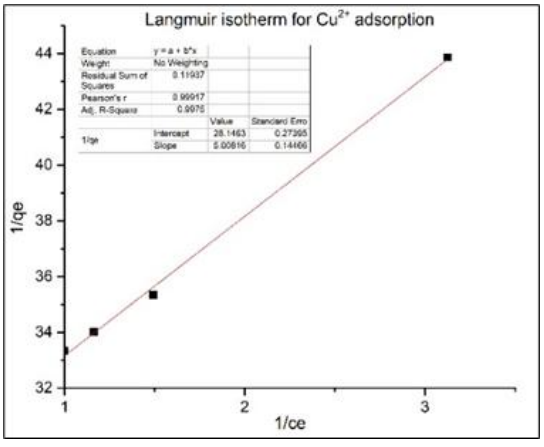

Figure 10: Langmuir isotherm for Cu^{2+} adsorption.

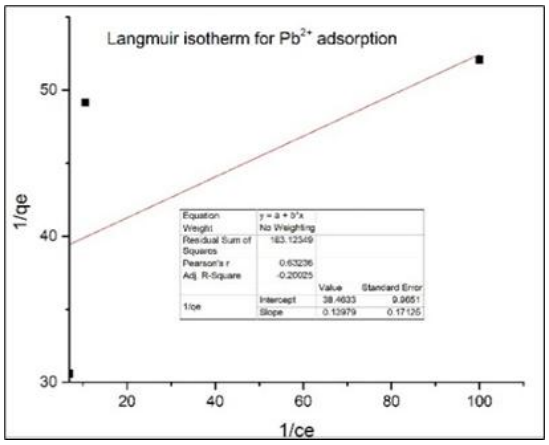

Figure 11: Langmuir isotherm for Pb^{2+} adsorption.

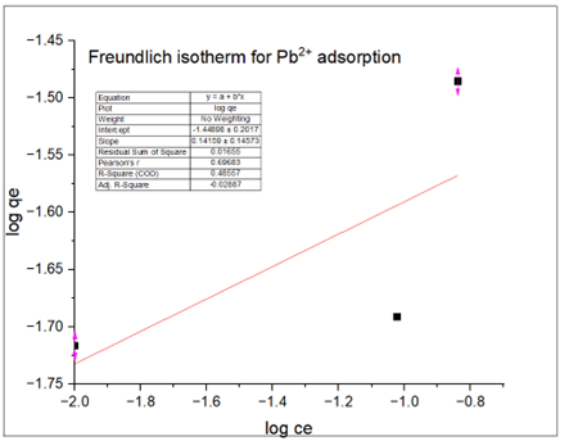

Figure 12: Freundlich isotherm for Cu^{2+} adsorption.

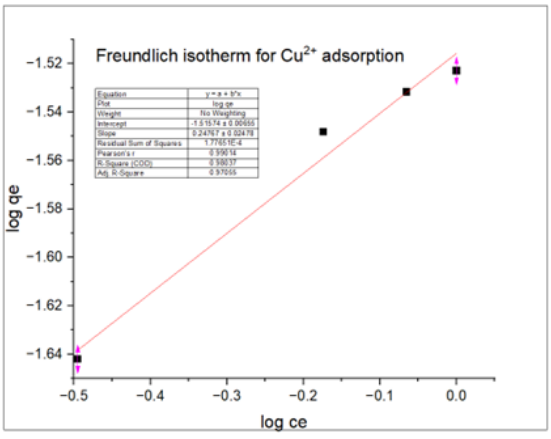

Figure 13: Freundlich isotherm for Pb^{2+} adsorption.

The summary of the analysis results is presented in Table 1

Table 1: Isotherm Parameters for Cu^{2+} and Pb^{2+} adsorption into APKS

Model	**ion**	**qmax (mg/g)**	**b (L/mg)**	**R^2**
Langmuir	Cu^{2+}	0.035	5.62	0.997
	Pb^{2+}	0.026	275.15	-0.200
	ion	**n**	**Kf**	**R^2**
Freundlich	Cu^{2+}	4	0.03	0.970
	Pb^{2+}	7	0.04	-0.03

According to Desta (2013), the n values for the Freundlich isotherm model, ranging between 1 and 10, indicate good adsorption. Therefore, according to the n-values obtained from this study, the adsorption process was suitable for both Cu^{2+} ions and Pb^{2+} ions concentrations. Furthermore, since the n-value was more significant than 1, the adsorption process was physical adsorptions (Al-Ghouti and Da'ana. 2020). When comparing the Langmuir and Freundlich adsorption constants and correlation coefficients (R^2) as presented in Table 1, results showed that both Langmuir and Freundlich adsorption isotherm was the best model for the adsorption of Cu^{2+} ions on APKS, with R^2 of 0.997 and 0.97, respectively. However, when comparing the Langmuir and Freundlich adsorption constants and correlation coefficients (R^2) for the Pb^{2+} adsorptions, the results showed that neither Freundlich nor Langmuir adoptions model was fit to describe the adsorption process for Pb^{2+} ions, the study recommends the testing of other adsorption isotherm models such as Brunauer-Emmett-Teller isotherm.

CONCLUSIONS

The results of the dosage effects show that a higher Cu^{2+} concentration adsorption capacity with 92% removal efficiency was attained at a dosage of 5 g/150 ml and a contact time of 20 minutes. On the other hand, at the dosage of 15 g, the higher the Pb^{2+} concentration's adsorption capacity was achieved with 98.6% removal efficiency. The findings on the initial concentrations demonstrated that the higher Cu^{2+}

concentration removal efficiency of 84.29 % was attained at an initial adsorbent dosage of 4.0 mg/L and an 80-minute contact time. The removal of 99.9% of the Pb^{2+} concentration was accomplished with an initial adsorbent dosage of 1.93 mg/l at the 20-minute contact time. With the column model, the APKS removed Pb^{2+} and Cu^{2+} concentrations with efficiencies of 98.4% and 73.6 %, respectively. In light of these findings, the study suggests that wastewater be treated with activated palm kernel shells to reduce Pb^{2+} and Cu^{2+} concentrations before it is discharged into the environment.

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Conflict of Interest

We declare no conflict of interest in this research work.

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