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Original Research Article

Kinetic and Isotherm Study of the Biosorption of Zinc (II) and Copper (II) Ions on Banana Peel Powder

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ABSTRACT

Utilization of agricultural waste as an alternative approach for removing heavy metals from industrial effluents has gained popularity. This study was carried out to investigate the effectiveness of banana peel powder (BPP) as an adsorbent for the removal of zinc and copper metal ions from aqueous solutions. Banana peels were dried, grounded and sieved to uniform sizes of 150µm powder. Biosorption experiments were carried out in batches for each metal at variable pH (3 – 8), adsorbent dosage (0.5 – 2 g/l) and contact time (10 – 90 min) using one parameter at-a-time approach. The highest removal of Zn (II) ions observed was 96 % at pH of 8, dosage of 1 g/l in 60 min. However, for Cu (II) ions, the highest was 71 % at pH of 6, dosage of 1 g/l in 60 min. The results consistently showed that banana peel is far more effective in removing zinc than copper. The equilibrium adsorptions of both zinc and copper were best described by the Langmuir isotherms with correlation coefficients of 0.96 and 0.98 respectively. The results further showed that adsorption of zinc followed pseudo-first-order kinetics while copper was best described with pseudo-second-order kinetics. The data of FT-IR spectrum confirmed the presence of ionisable functional groups (i.e., amino, carboxyl, and hydroxyl), which were able to interact with zinc and copper metal ions. These results suggest that banana peel can be used as an effective adsorbent for removing heavy metals such as zinc and copper ions from aqueous solution.

1. INTRODUCTION

Increased agricultural waste and aqueous solution contamination by heavy metals has become a global trouble especially for developing countries. The growing demand and utilization of metals and chemicals in industries such as tanneries, metal plating, mining, fertilizer production, petroleum refining, paint manufacture, pesticides, pigment manufacture, and printing and photographic industries has led to the generation of substantial amounts of wastewater containing high concentrations of harmful heavy metals like zinc, copper, cadmium, aluminum, and nickel (Sharma & Bhattacharyya, 2005; Zhang et al., 2009; Zvinowanda et al, 2010; Babarinde, 2011).

In contrast to organic pollutants, heavy metals are non-biodegradable and remain within the environment over long time (Feng et al., 2010). Zinc and copper are ubiquitous metal ions in soil and aquatic environments (Zhang et al., 2013). At low concentrations, they are important nutrients for the body. On the other hand, they cause serious risk and threaten both human health and the environmental when present above their permissible limits (Yilmaz & Tugrul, 2022; Danial & Dardir, 2023).

Several methods have been applied to remove excessive heavy metals from aqueous solutions such as chemical precipitation, coagulation, flocculation, ion exchange, reverse osmosis (Esalah et al., 2000; Cardoso et al., 2004; Altıntıg et al., 2021; Badmus et al., 2021; Saleh, 2021; Yilmaz & Tugrul, 2022). However, their operational expenses limit the use of these methods. Biosorption has emerged as an attractive substitute technique due to large availability, low cost, operation ease and high heavy metal adsorption efficiency of the adsorbents (Zhang et al., 2013; Jalija et al., 2019; Oyewole et al., 2019; Yilmaz & Tugrul, 2022).

Different agricultural waste, being natural, abundant and economic, have been utilized for the removal heavy metals in wastewaters and to decrease the pollution grade. Most researchers have tried rice husk, tea leaves, corncobs, coconut husk, barley husks, peanut hull, tangerine peels, watermelon peel, wheat bran, eggshell, banana peel, neem leaves, etc. (Ahluwalia, 2005; Saeed et al., 2005; De Gisi et al., 2016; Mathew et al., 2016; Singh et al., 2020). Banana is one of the very extensively cultivated tropical fruits, planted over 130 countries. More than 25% of the fruit is dry mass and the rest is water (Ali 2017; Vilardi et al., 2018). Banana peels have high content of hydroxyl and carboxyl groups (Zhang et al., 2013; Arifiyana & Devianti, 2021) therefore can be used as a biosorbent for heavy metal adsorption from aqueous solutions. The large amount of discarded banana peel poses significant challenges for disposal and represents a significant wastage of resources.

In this work, banana peel, generally regarded as nuisance waste product, was used as an alternative economic biosorbent for zinc and copper adsorption from aqueous solution. The work aimed to compare the adsorption capacities, kinetics and isotherms of zinc and copper sorption on banana peels. The SEM and FT-IR spectrum of the adsorbent were also analyzed.

2. MATERIALS AND METHODS

2.1. Reagents and Apparatus

The reagents and main apparatus used include: analytical grade zinc chloride (98 %, Aigma Aldrich, Germany), analytical grade copper chloride (97%, Aigma Aldrich, Germany), reagent grade sodium hydroxide (98 %, Aigma Aldrich, Germany), hydrochloric acid (Merck, Darmstadt, Germany), magnetic stirrer (TOP-SH2, China), Atomic Adsorption Spectrometry (Perkin Elmer AAnalyst 200, Germany), Scanning Electron Microscope (JSM-7610F Schottky Field Emission SEM, Japan), Fourier Transform Infrared Spectroscopy (JASCO FTIR 6100, Japan), Cutting Mill (KM-1500, UK), Floc Tester Unit (Edibon PEF, Spain), pH meter (JENWAY 3510, USA), Oven (Genlab MINO/50, UK), Weighing Balance (Ohaus SP202, Scoutt Pro, USA). Distilled water was produced using water distiller (SZ-96 Mon Scientific, Nigeria). Other apparatus used were stopwatch and glassware.

2.2. Preparation of Biosorbent

Fresh banana peels were collected from the Center for Dryland Agriculture (CDA) Bayero University, Kano, Nigeria. The peels were washed with distilled water to remove dirt and foreign particles. It first was dried under sun for 6 hr and then further dried in an oven at 105 ℃ for 24 hr. The dried peels were crushed, ground and sieved to obtained fine powder with an average size of approximately 150 μm. Finally, the banana peel powder (BPP) was stored in an airtight polyethylene bags at room temperature prior to usage

2.3. Biosorbent Characterization

The morphological and surface structure of the biosorbent was analyzed using scanning electron microscope at different magnifications while, the functional groups present in the biosorbent was determined using Fourier transform infrared (FT-IR) spectroscopy with scanning range of 4000 – 400 cm^{-1}.

2.4. Preparation of Metal Solutions

A stock solution of Zn (II) ions was prepared by dissolving the 1 g of Zn (II) chloride salts in 1000 ml of distilled water. Another stock solution containing 1g/l Cu (II) ions was prepared by dissolving 1 g of Cu (II) chloride salts in 1000 ml of distilled water. Experimental solutions (100 mg/l) for each metal were prepared by diluting the respective stock solution ten times (10x) with distilled water. Diluted HCl and NaOH solution were used to adjust the pH of each solution.

2.5. Batch Experimental Method

Batch experiments were conducted to investigate the effects of process parameters such as pH, biosorbent dosage, and contact time on the biosorption of each of the metal ions (zinc and copper). The experiments were carried out by varying *one parameter at a time* while keeping the others constant. The experiments for zinc adsorption were as follows: Initially, 250 ml of a solution containing 100 mg/l of Zn (II) ions was prepared in a conical flask. The pH was adjusted to a desirable value (3 – 9). Then appropriate amount of the BPP (to give a dosage of 0.5 – 2 g/l) was added and the mixture was agitated using a floc tester unit for 60 min at 200 rpm at room temperature. The container was allowed to settle for 30 min and the residual metal concentration in the supernatant was determined using AAS. The same procedure was applied for the copper adsorption experiments. The percentage of metal ions removal and adsorption capacity of the biosorbent (metal ion uptake) was calculated using Equations 1 and 2 respectively.

$$Removal\ (\%) = \frac{(C_i - C_f)}{C_i} \times 100 \quad (1)$$

$$q_e = \frac{V(C_i - C_f)}{M} \quad (2)$$

Where C_i (mg/L) is the initial adsorbate concentration, C_f (mg/L) is the final adsorbate concentration, q_e (mg/g) is the amount of metal ion adsorbed per unit mass of adsorbent, V (L) is the solution's volume and M (g) is the amount of the biosorbent.

2.6. Determination of Biosorption Isotherms

The Langmuir model assumes monolayer biosorption on identical and energetically equivalent active sites without any interaction between adsorbed molecules (Sutirman et al., 2018).

$$\frac{C_e}{q_e} = \frac{1}{kq_m} + \frac{C_e}{q_m} \quad (3)$$

Where q_e is adsorption capacity at equillibrium (mg/g), q_m is the maximum capacity at equillibrium (mg/g), K is the constant related to the affinity of the binding site (l/mg), and Ce is the adsorbate equilibrium concentration (mg/L).

While the Freundlich isotherm describes the multilayer biosorption with the non-uniform distribution of sorption sites over the heterogeneous surface along with interactions between adsorbed molecules (Sutirman et al., 2018).

$$log q_e = log K + \frac{1}{n} \log C_e \quad (4)$$

Where q_e is adsorption capacity at equilibrium (mg/g), K and n are Freundlich constants related to adsorbent adsorption capacity and affinity of the binding site respectively and C_e is the adsorbate equilibrium concentration (mg/L).

2.7. Biosorption Kinetics

The kinetics of the metals adsorption was studied using kinetic models which are pseudo-first order and pseudo-second order using Equations 5 and 6 respectivly (Zhang et al., 2013) .

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (5)$$

$$\frac{t}{q_t} = \frac{1}{k_2 {q_e}^2} + \frac{1}{q_e} t \quad (6)$$

Where, q_e is adsorption capacity at equillibrium (mg/g), q_t (mg/g) is the adsorption capacity at any given time t (s), k_1 (s^{-1}) and k_2 (s^{-1}) are the equilibrium rate constant of pseudo-first-order and pseudo-second-order respectively.

3. RESULTS AND DISCUSSION

3.1. Characterization of the Biosorbent

The biosorbent active sites involve in the metal uptake, the biosorbent has a porous morphology structure as indicated in Figure 1. The banana peel has a vast heterogenous rough surface with a honey comb-like pores, and the particles has an irregular shape with a micro-rough texture on their surface, which can enhance zinc and copper adhesion (Arunakumara et al., 2013; Mondal, 2017). The biosorbent have showed a rough surface area with irregular crystal structure and a greater surface area per field. Figure 1 indicates the biosorbent surface at range of 500-1500× magnifications. It showed BPP is enriched with many pores and are well distributed on the surface. The pores obviously indicate presence of many active sites on the surface of the biosorbent.

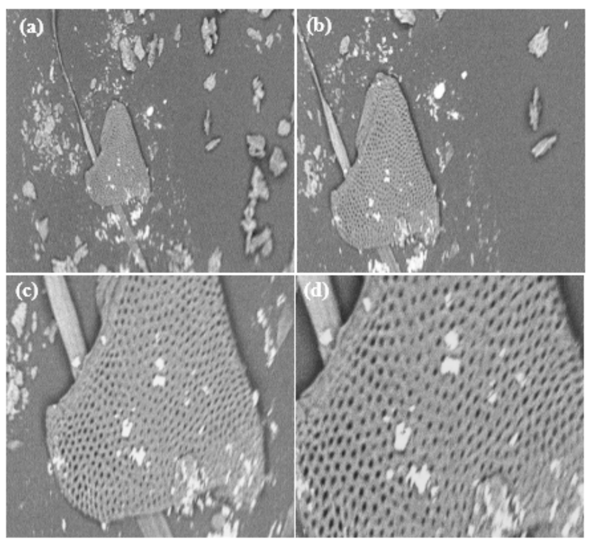


Figure 1: SEM of biosorbent (a) magnified 300× (b) 500× (c) 1000× (d) 1500×

FITR spectra of the biosorbent was used to examine the functional groups present on the BPP structure, and possible interaction between the heavy metal ions and the functional groups of biosorbent as indicated in Figure 2. The adsorption band at about 3,272 cm^{-1} demonstrated O-H stretching vibration of hydrogen-bonded carboxylic acid, phenol and alcohols. The band at about 2,922 cm^{-1} might be due to antisymmetric and symmetric stretching of C–H bond of methyl and methylene groups and aliphatic acids. The bands at 1,599– 1,734 cm^{-1} demonstrated carbonyl C=O groups (carboxylic acid, acetate groups, ketone, aldehyde) and the stretching vibration bond of (C=O) and (C=C). The band at about 1,028 cm^{-1} belongs to C-O stretching vibrations in alcohols, phenols, ether and ester groups. Therefore, these results have indicated that the banana peel contained lipids, protein and carbohydrates. These functional groups have the ability bind heavy metals through ionic, weak electrostatic interactions, and Van der Waals forces (Shakoor & Nasar 2016; Khan et al., 2017; Vilardi et al., 2018)

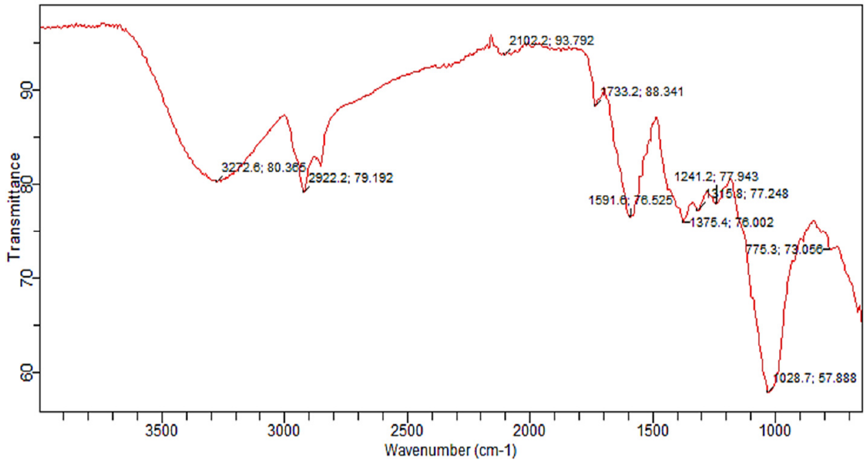
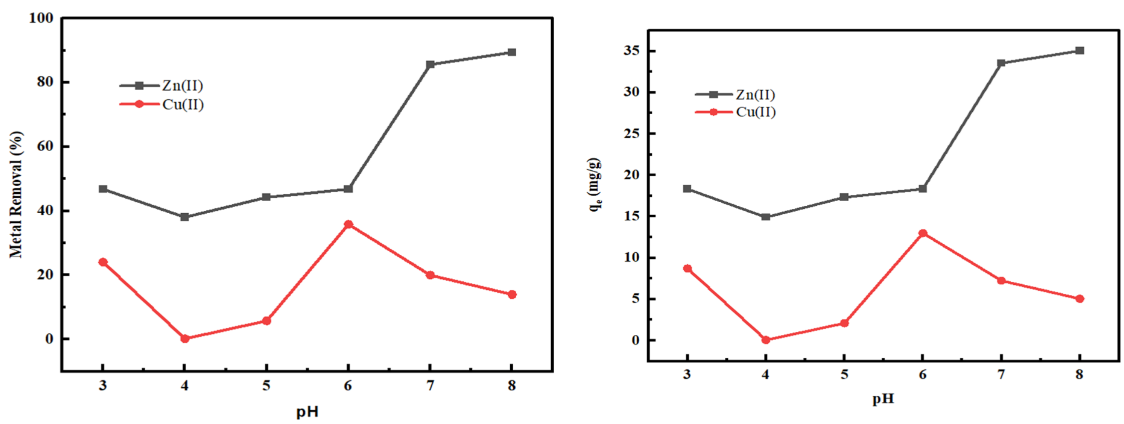


Figure 2: FT-IR spectra of BPP

3.2. Effect of pH

The effect of pH was investigated between the range of 3 to 8 at constant temperature and agitation speed of 200 rpm, BPP dosage of 1g/l for 60 min. The pH level plays a crucial role in determining the adsorption of metals from an aqueous solution. It influences the surface characteristics of adsorbents, the ionic state of functional groups, and the behavior of metal species (Seleman et al., 2023). Therefore, the adsorption of Zn^{2+} and Cu^{2+} ions are more favored in basic and acidic medium. It can be seen from Figure 3 that the percentage removal and adsorption capacity increased from pH of 4 to 6 for both zinc and copper..



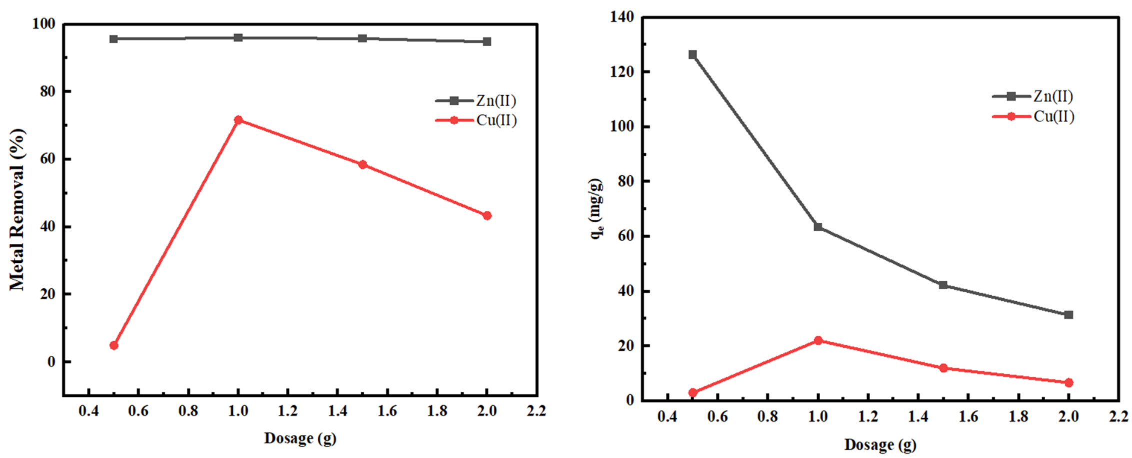
(a) (b)

Figure 3: Effect of pH on (a) percentage removal (b) adsorption capacity of the Zn (II) and Cu (II) ions

However, highest sorption of zinc was achieved at pH of 8 (slightly basic) with 90 % and adsorption capacity of 35.05 mg/g. In contrast, only 15 % of copper was removed at pH of 8. The highest removal of copper was achieved at pH of 6 (slightly acidic) with 36 % and adsorption capacity of 13 mg/g. According to Keawkim & Khamthip (2018), under highly acidic conditions, the primary functional groups responsible for binding sites on the sorbents are less ionized. As a result, the positively charged metal ions are not attracted to the negatively charged functional groups present on the surface of the adsorbent. Furthermore, the metal ions are unable to attach to the active sites on the surface because these sites are predominantly occupied by proton ions (H^+). The results obtained in this study for both zinc and copper are in agreement with Tahirudeen et al. (2023) and Hossain et al. (2012) who reported pH of 8 and 6 as the optimum values for zinc and copper removals respectively. It was also observed that at higher pH conditions, the efficiency of the copper removal decreased compared to the optimal condition while the reverse was observed in the case of zinc

3.3. Effect Biosorbent Dosage

The effect of absorbent dosage was investigated between the range of 0.5 to 2 g/l at constant temperature and pH of 8 while maintaining an agitation speed of 200 rpm for 60 min. The effect of the adsorbent-to-solution ratio dictates the adsorption capacity of the adsorbent during batch sorption (Murugan et al., 2010). As observed in Figure 4, more than 95 % of zinc was removed even at lower dosage of 0.5 g/l. The same removal percentage was observed at 2 g/l. This suggests that active sites of the BPP are many and have great affinity for Zn metal ions. On the contrary, only 5 % of copper was removed at lower dosage of 0.5 g/l. However, increasing the dosage to 1 g/l allowed for the removal of about 70% of copper. This phenomenon can be understood by considering the increased surface area of the adsorbents, resulting in a higher number of accessible active sites in comparison to the metals concentrations (Oyebamiji et al., 2011). However, removal of copper declined as the dosage increases above 1g/l, this can be attributed to the fine nature of BPP which agglomerates at higher dosage leading to the blockage of the available active sites (Rasheed et al., 2014). The highest zinc adsorption capacity was found to be 126 mg/g at 0.5g/l BPP dosage. While the copper adsorption capacity was found to be 22 mg/g at 1g BPP dosage as shown in Figure 4.

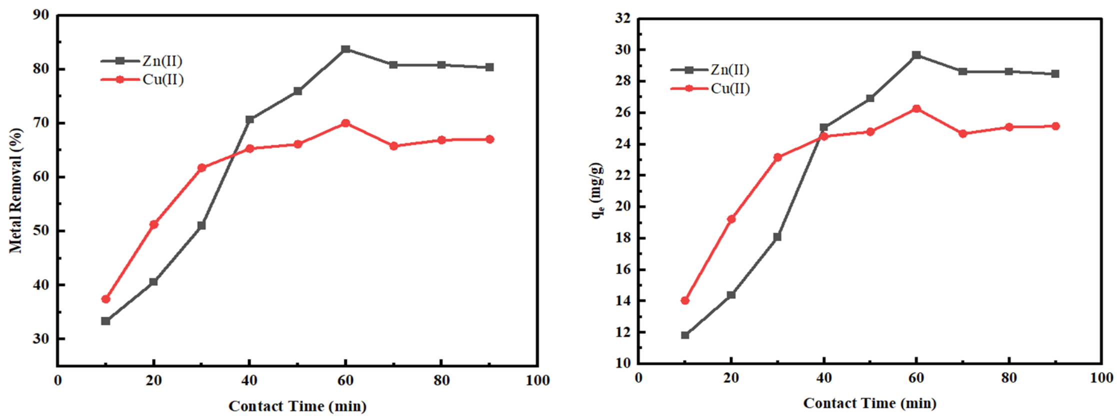


(a) (b)

Figure 4: Effect of BPP dosage on (a) percentage removal (b) adsorption capacity of Zn (II) and Cu (II) ions

3.4. Effect of Contact Time

The effect of contact time was investigated in the range of 10 to 90 min at constant temperature and pH of 8 and dosage of 1g/l while maintaining an agitation speed of 200 rpm. Figure 5 shows the amount of zinc and copper ions adsorbed on the BPP as a function of time. It can be observed that zinc percentage removal and adsorption capacity are higher than that of copper. The rate of the metals removal was rapid for the first 40 min, and then reached equilibrium after 60 min. This suggests that the initial rapid adsorption was due to the presence of the large number of vacant active binding sites on the banana peel. However, after a while, the available binding sites becomes saturated which makes them difficult to bind with metals ions due to repulsive forces between the metals already bound to the solid surface and those in the liquid phase (Achak et al., 2009). Additionally, the momentum for mass transfer between the liquid and solid phase in a water-based adsorption system reduces. Moreover, the metal ions need to penetrate deeper into the pores for binding, facing higher resistance. This resistance prolongs the adsorption process in the subsequent adsorption stages (Srivastava et al., 2006).

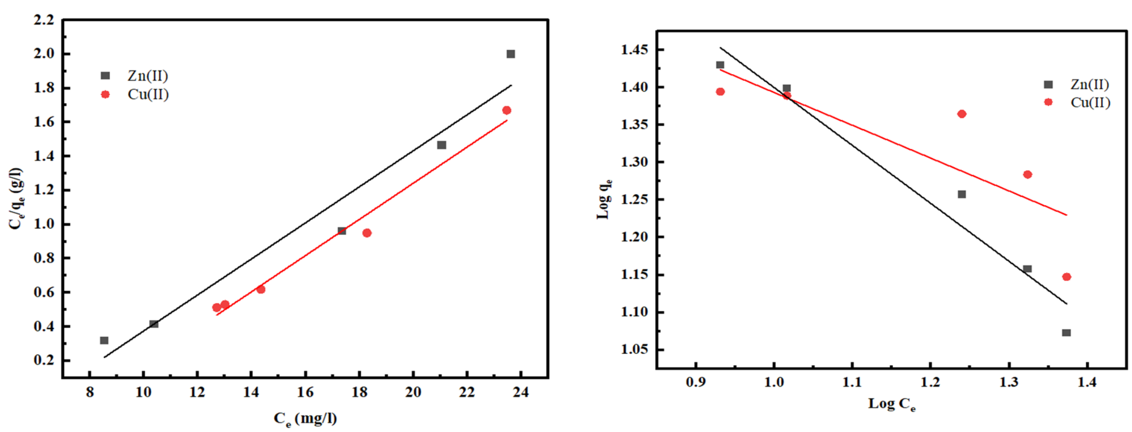


(a) (b)

Figure 5: Effect of contact time on (a) percentage removal (b) adsorption capacity of Zn (II) and Cu (II) ions

3.5. Biosorption Isotherms

In this study, the equilibrium adsorption data were fitted using the Langmuir and Freundlich models. This was used to understand the interaction between metal ions and BPP. Figure 6 depicts the graphical representations of linear isotherm models for the adsorption of Zn (II) and Cu (II) ions onto BPP. Additionally, Table 1 provides the estimated isotherm parameters.



(a) (b)

Figure 6: Biosorption isotherms (a) Langmuir (b) Freundlich of the Zn (II) and Cu (II) ions

Table 1: Biosorption isotherms parameters of Zn (II) and Cu (II) removal using BPP

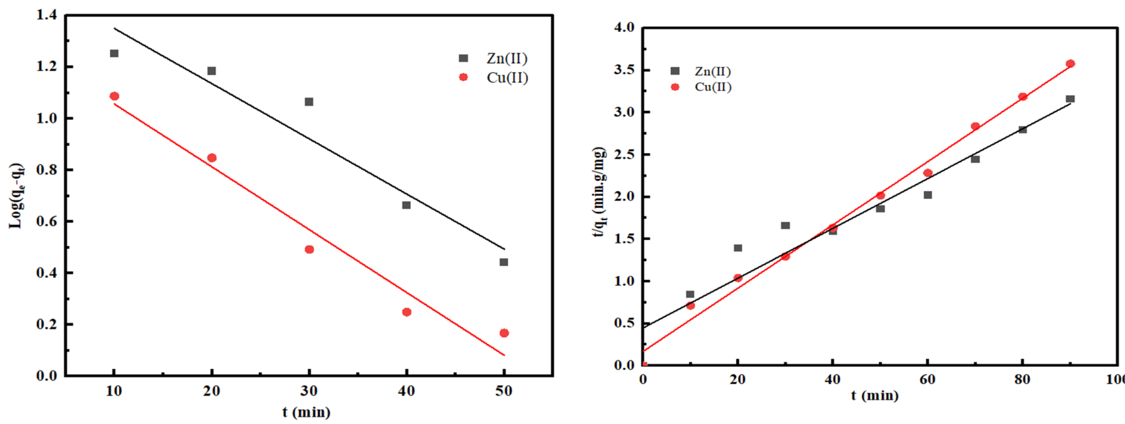
Metal ions	Langmuir			Freundlich		
	q_m (mg/g)	b (mg/g)	R^2	k (mg/g)$(1/g)^{1/n}$	n	R^2
Zn(II)	9.450	-0.155	0.960	148.730	-1.291	0.957
Cu(II)	9.416	-0.121	0.980	255.682	-1.101	0.978

According to the Langmuir model, a monolayer adsorption phenomenon occurs on a homogeneous surface characterized by limited adsorption sites and low interactions between the adsorbed molecules. On the other hand, the Freundlich model proposes that molecules adsorbed to the heterogenous surfaces of adsorbents at distinct binding sites, each with its own adsorption energy (Chen et al., 2014; Kumari et al., 2022). Figure 6 shows that the Langmuir models fits the data better than the Freundlich models for Zn (II) and Cu (II) removal using BPP. This conclusion is supported by the Langmuir model's high R^2 value compared to the

Freundlich model as shown in Table 1. The Langmuir isotherm indicates the adsorbent's surface homogeneity. This further suggests uniformity of binding sites on the adsorbent surface, where each adsorbate molecule/adsorbent have the same activation energy for adsorption (Jawad et al., 2018). More notably, it suggests that the adsorbent can accommodate a greater amount of Zn (II), exhibiting its heightened inclination for Zn (II) in comparison to Cu (II).

3.6. Biosorption Kinetics

Adsorption kinetics, which is one of the important characteristics defining the efficiency of biosorption, described the metal uptake rate. In this study, pseudo-first order and pseudo-second order models were used to predict Zn (II) and Cu (II) adsorption kinetics on the banana peel surface. Figure 7 shows the plot for zinc and copper ions respectively. The rate constants and correlation coefficients of kinetics are presented in Table 2. It can be observed from Figure 7 that the linear plot for Zn (II) ions is better described and fitted by pseudo-first-order kinetics with higher value of reaction constant (K_1), and higher correlation (R^2) between experimental and measured q_e. While Cu (II) ions removal was better fitted by pseudo-second-order kinetics. The results suggest that the zinc and copper adsorption on the banana peel follows pseudo-first-order and pseudo-second-order kinetics respectively. It was also observed here that the equilibrium adsorption capacity of zinc was greater than that of copper as consistently seen throughout the study.



(a) (b)

Figure 7: Biosorption kinetic models (a) Pseudo-first-order (b) Pseudo-second-order of the Zn (II) and Cu (II) ions

Table 2: Biosorption kinetics parameters of Zn (II) and Cu (II) removal using BPP

Metal ions	Pseudo-first-order				Pseudo-second-order		
	$q_{e\ exp}$ (mg/g)	K_1 (min^{-1})	$q_{e\ cal}$ (mg/g)	R^2	K_2 (min^{-1})	$q_{e\ cal}$ (mg/g)	R^2
Zn(II)	29.677	0.049	36.590	0.923	6.205E-04	44.840	0.831
Cu(II)	26.269	0.056	19.950	0.965	2.702E-03	31.250	0.996

4. CONCLUSION

The adsorption of zinc and copper metal ions by banana peel powder was examined in this study. The results showed adsorption dependence for both metals on pH, BPP dosage and contact time. The highest removal of Zn (II) ions observed was 96 % at pH of 8, dosage of 1 g/l for 60 min. However, 71 % was the highest observed for Cu (II) at pH of 6, dosage of 1 g/l in 60 min. The results consistently showed that banana peel is far more effective in removing zinc than copper. The equilibrium adsorptions of both zinc and copper were best described by the Langmuir isotherms with correlation coefficients of 0.96 and 0.98 respectively. The results suggest that adsorption of zinc was better described by pseudo-first-order kinetics while copper was best fitted by pseudo-second-order kinetics. The FT-IR data spectrum showed the presence of various functional groups (including amino, carboxyl, and hydroxyl) which are able to interact with both zinc and copper ions. This study shows that agro-waste, such as banana peels, can be considered as an effective biosorbent for the removal of heavy metals such as zinc and copper from aqueous solutions.

5. ACKNOWLEDGMENT

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6. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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