

Article

Alginate-Immobilized *Arachis hypogaea* Hull: Characterization, Kinetics, and Sustainable Biosorption Removal of Some Toxic Metals from Municipal Wastewater

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Abstract

Sustainable development is seriously hampered by environmental contamination. Heavy metals are perhaps the most significant of these silent pollutants because of the poisoning they inflict on different ecosystems, particularly water systems, as well as the direct threat they pose to the safety of such systems and the ensuing health issues that affect people and other living things. The goal of the current work is to ascertain how effective peanut (*Arachis hypogaea*) hulls are as easily accessible and reasonably priced adsorbent materials for the ion adsorption of various heavy metals, including lead, copper, zinc, and cadmium. A series of variables, such as pH, temperature, retention time, shape, quantity of adsorbent and particle size, were investigated to reach the optimal conditions for the biosorption process, which were achieved at pH 6 (close to pH-zero charge), a temperature of 40 °C, a retention time of 60 min, an adsorbent dose of 5 g/L, and a particle diameter of 0.1 mm. Experiments showed that lead had the highest treatment rate, approximately 95%, followed by zinc, copper, and cadmium. The results were confirmed and validated using SEM analysis to identify the morphology of the hulls before and after treatment, FTIR spectroscopy to investigate the functional groups responsible for binding to the metal ions, and further supported by kinetic isotherms of the adsorption behavior, which matched the Langmuir and Freundlich models. Nevertheless, the results of the batch experiment were employed by designing a laboratory treatment unit for municipal wastewater containing beads of alginate-immobilized *Arachis hypogaea* hulls. The designed laboratory unit was effective in treating unacceptable levels of heavy metals in some municipal wastewater from the local drainages of Ramadi City, Iraq. Overall, the study's findings suggested that peanut hulls could be used to mitigate toxic metal pollution in an eco-friendly manner that guarantees the accomplishment of the Sustainable Development Goals pertaining to future generations' right to the safe use of water resources.

Keywords: biosorption; peanut hulls; heavy metals; alginate immobilization; kinetics; wastewater



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1. Introduction

Over the last three decades, there has been increasing worldwide apprehension regarding the health impacts of environmental contamination or pollution, more specifically the

global burden of diseases. Approximately a quarter of the total disease burden in humans can be attributed to environmental exposures that have occurred over long periods. Most diseases related to pollution are not easily diagnosed, mostly acquired during childhood, and may become evident in later life [1]. A major environmental concern is the presence of heavy metals in aqueous streams and drains; this is due to the release of untreated effluent containing metals into water bodies. Heavy metals such as lead, zinc, copper, and cadmium are common in industrial and/or municipal wastewater. They pose a risk to the aquatic ecosystem and can potentially affect human health [2,3].

Lead, for example, is a common pollutant. It is very dangerous even in low doses. It is one of the main heavy metals that can cause problems in the body, being linked to neurological and renal problems as well as heart disease [4]. The current Environmental Protection Agency (EPA) and World Health Organization (WHO) standards for lead in drinking water are 0.05 mg/L and 10 µg/L, respectively [5]. Some possible sources of lead in industrial waste are battery manufacturing, basic steel, paper, pulp, metal plating, leather tanning, mining, and fertilizer industries [6]. However, there are various industrial applications of lead and zinc, such as in electroplating, dry batteries, and in the forming of different alloys. It may also be mixed with chlorine, oxygen, and sulfur, where such compounds are used in the industry of paints, ceramics, medicines, and ointments [7]. Zinc is also a constituent of enzymes and nucleic acids and therefore has a basic function in body building and immune defense; however, large amounts (above the requirements) are toxic and cause symptoms like nausea, vomiting, and damage to the pancreas [8]. Similarly, copper is one of the essential elements required for the growth of plants, animals, and humans, because their normal metabolic processes need very small amounts. Yet, overexposure leads to anemia and digestive disorders, shown through vomiting and diarrhea [9]. In addition, cadmium occurs mainly in the Earth's crust and is also present in industry as an inevitable by-product in the extraction of zinc, lead, and copper. Because it is present in manures and insecticides, it mainly enters the environment via soil after spraying. A huge amount of cadmium can be released into the ecosystem naturally, at approximately 25,000 t per year. About half of it is released into rivers by rock weathering, and some of it can be released directly into the air by forest fires and volcanoes. The rest is released due to human activities, like manufacturing and smoking [10,11]. Cadmium can transport to the liver through the blood. Hence, it is bound to protein to form complex compounds that transport to the kidney where they are accumulated and damage the filtering mechanism. This may result in the secretion of important sugars and proteins from the body and cause further kidney damage [12]. In view of this toxicity and the need to meet regulatory standards for safe discharge, it is necessary to remove such metals from wastewaters/effluents before they are released into the environment.

Different techniques have been developed for the elimination of heavy metal ions from industrial wastewater, including chemical precipitation, ion exchange, coagulation, electrolysis, reverse osmosis processes, electro-flotation, etc. [13,14]. Most, however, exhibit some drawbacks, such as a cumbersome treatment process, high cost, and energy use. Therefore, the quest for a low-cost and competitive method is highly recommended.

Adsorption is much more preferable to other techniques for heavy metal removal from polluted waters because of its simplicity and low cost of operation [15]. Even though activated carbon is the most promising adsorbent for the adsorption process, having a large surface area and high adsorption capacity, it is also very expensive, operation costs are high, and it has to be regenerated after each adsorption cycle [16]. Therefore, an alternate low-cost sorbent material is urgently required to replace high-cost activated carbon for water and wastewater treatment. In the last two decades, several low-cost materials have been tested for their potential to adsorb heavy metals. One such material is agricultural

by-products; for example, walnut hulls [17], rice husk [18], and plant leaves are gaining much attention from researchers for wastewater treatment due to their wide availability and low cost [19]. One of these low-cost sorbents that is particularly suited to adsorption is *Arachis hypogaea* hulls or peanut hulls, which exhibit a large metal-binding capacity. This material is an agricultural crop residue that cannot be used as animal feeds and is usually burnt or dumped as municipal waste, which in both ways increases pollution. Peanut hulls consist of many components, namely cellulose, hemicellulose, and lignin, along with other minor constituents such as oil, protein, etc. [20]. Metal ion uptake by peanut hulls may involve complexation, coordination, chelation, ion exchange, and adsorption. The mechanism of binding metal ions via this adsorbent may depend on the species and ionic charges of the metal ions, the chemical compositions of the metal ion solution, and other external environmental factors such as pH and temperature.

To assess the biosorption of Pb, Zn, Cu, and Cd from aqueous solution by the hulls of *Arachis hypogaea*, the following specific parameters were determined: effect of pH, retention time, temperature, size of hull powder, and dose. Equilibrium data were analyzed using the Langmuir and Freundlich isotherm models. Possible mechanisms involved in the removal of heavy metal ions were investigated by FTIR and SEM. A laboratory treatment unit was formed that will include the results of biomass immobilized beads for the establishment of a field processing system. The aim of this study lies in finding a highly efficient material for treating drinking water pollution with heavy metals in an environmentally friendly and low-cost manner, without generating polluting by-products, and for eliminating agricultural waste burdening the environment, thus contributing to achieving sustainable development goals.

2. Materials and Methods

2.1. Preparation of Biosorbent

Arachis hypogaea hulls were obtained from the local markets of Ramadi City, Iraq. The hulls were then washed with double-distilled water to make them free from any impurities and to remove any adhering residues. After cleaning, the hulls were prepared by two different methods: in the first method, they were dried thoroughly and then used wholly or cut into pieces of different sizes of dried peanut hull (PH); in the second method, the dried hulls were grinded to a fine powder and sieved using stainless steel sieves with pore sizes between 0.106 mm and 2 mm to obtain dried peanut hull powder (PHP). Figure 1 illustrates the prepared biosorbents. The biosorbent was kept in a dry clean container to avoid moisture pick-up and used in adsorption experiments for studies on the removal of metal ions [19].

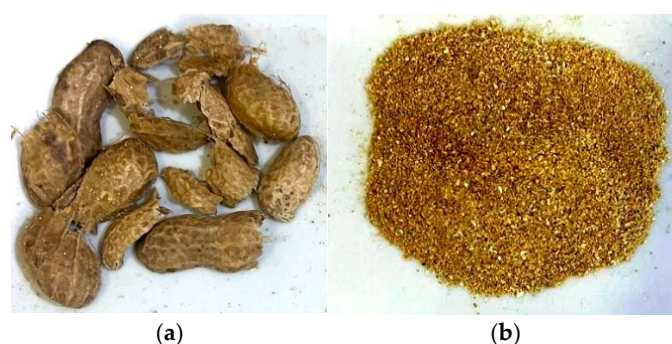


Figure 1. Biomass of adsorption: (a) peanut hull (PH), (b) peanut hull powder (PHP).

2.2. Description of *Arachis hypogaea*, SEM, and FTIR Analysis

The pH_{pzc} of the *Arachis hypogaea* PHP biosorbent was determined by the pH drift method. A series of 0.1 M NaCl (Fluka, Buchs, Switzerland) solutions was prepared in 100 mL conical flasks. The initial pH (pH_i) of each solution was adjusted within the range of 2–12 using 0.1 M HCl or NaOH (Fluka, Buchs, Switzerland) and measured by a pH meter (Thermo Scientific, Orion Star A211, Jakarta, Indonesia). The volume was made up to mark, i.e., 25 mL. To all the flasks, 0.5 g of *Arachis hypogaea* PHP was added and then allowed to equilibrate by shaking at 120 rpm for 24 h. The final pH (pH_f) was recorded. The difference between initial and final pH values ($\Delta pH = pH_i - pH_f$) was plotted against pH_i . The pH_{pzc} was determined at the point where $\Delta pH = 0$ on the x -axis [18].

The surface morphology of raw and metal-loaded PHP biosorbent was examined using scanning electron microscopy (SEM) (Tescan–Vega III, Brno, Czech Republic). For the adsorption experiment, a multi-metal solution containing 100 mg/L of each ion (Fluka, Buchs-Switzerland) of lead (Pb), copper (Cu), zinc (Zn), and cadmium (Cd) was prepared. One gram of *Arachis hypogaea* powder (PHP) was added to the solution and allowed to equilibrate for 2 h prior to SEM analysis [17].

The functional groups that participate in binding with metal were determined by using Fourier Transform Infrared (FTIR) spectroscopy (ABB Spectrolab, MB3000, Quebec, QC, Canada). The biosorbent samples were dried first and then mixed with potassium bromide (KBr) (Fluka, Buchs, Switzerland) in the ratio of 1:10. The mixture was ground finely and pressed into pellets. Spectra were recorded after background correction using pure KBr, and all results were put up on a uniform absorbance scale [17–19].

2.3. Experiments of Batch Biosorption

The biosorption performance of PHP biomass was evaluated in batch adsorption experiments conducted under controlled laboratory conditions. A multi-metal aqueous solution (100 mL working volume) was prepared with Pb^{2+} , Zn^{2+} , Cu^{2+} , and Cd^{2+} ions (Fluka, Buchs, Switzerland), each at an initial concentration of 100 mg/L, and then the solution was transferred into 250 mL volumetric flasks for further processing.

Biosorption conditions were optimized through preliminary experiments. This included evaluating the biosorbent form and physical characteristics of *Arachis hypogaea* biomass in both PH and PHP forms. In each experiment, 0.5 g of the biosorbent was added to 100 mL of metal solution. The flasks were then placed in a shaking incubator (LabTech, LSI-3016A, Daihan, Republic of Korea) at 120 rpm and 30 °C for 60 min at neutral pH. The solution was then adjusted to the desired pH using 0.1 M NaOH or 0.1 M HCl.

Additional optimization of biosorption parameters was carried out by studying the operational variables, namely initial pH of 3–8, temperature of 30 °C as control and 10–60 °C for thermal studies, contact time of 1–120 min, biosorbent dosage of 1–20 g/L, and particle size range from 0.106 to 2 mm. In particular, an optimal particle size effect was further considered. However, to ensure consistency in comparison, a fixed biosorbent dosage of 10 g/L was used. The value of optimum pH obtained from these sets of experiments was used in the subsequent adsorption studies. Additionally, a blank control solution that contains the same metal ions without PHP biosorbent was prepared under the same experimental conditions to serve as a reference for the evaluation of adsorption efficiency. After the biosorption process, all samples were centrifuged at 3000 rpm for 40 min at room temperature using Hettich–EBA 200S centrifuge, Tuttlingen, Germany. Then, they were subjected to filtration through filter paper of 45 µm to remove suspended particles. Residual concentrations of heavy metal ions in the aqueous phase were determined by Flame Atomic Absorption Spectrophotometry (FAAS) (AA-7000, Shimadzu, Kyoto, Japan) [17,18].

All experimental processes were conducted in a similar and repeatable manner to ensure the validity of the results and establish the best conditions for maximum biosorption efficiency of *Arachis hypogaea* PHP biomass. The overall removal efficiency was calculated by the standard adsorption efficiency equation, which relies on calculating the percentage of the treatment process based on the original concentration of the metal ion and the final concentration after the treatment process. These procedures were used in all the subsequent experiments to find the best efficient biosorption process. The overall efficiency of the biosorption process was measured using Equation (1) and the quantity adsorbed at equilibrium (qe) in mg/g was measured using Equation (2) [17]:

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

$$qe = (C_i - C_f) \times \frac{V}{W} \quad (2)$$

where (C_i) and (C_f) are the initial and final concentrations of metal, respectively. V (L) represents the volume of the heavy metal solution, while W (g) represents the weight of the PHP. However, to guarantee the results accuracy, all mensuration were carried out in triplicates, and the mean values were provided [18].

2.4. Biosorption Isotherms

The Langmuir and Freundlich isotherm models were used to describe the interaction between adsorbate species and the biosorbent surface in fitting the experimental equilibrium data and to provide insight into the adsorption mechanism. The above analysis was carried out to predict the equilibrium behavior of heavy metal ion adsorption onto *Arachis hypogaea* biosorbent. However, Langmuir defined gas adsorption on a solid interface as being analogous to gas-phase condensation on a liquid surface, wherein inelastic collisions result in a “lag” or residence time at the point of impact. The model assumed equal energy monolayer sites with no interaction between adsorbed molecules [20].

Further, the model examined only low to high pressures. These assumptions might not hold due to increasing pressure and heterogeneity in the rock fabric of hydrocarbon deposits. Therefore, the Langmuir model’s prediction accuracy might decrease. Experimental data is usually plotted to help indicate the nature of adsorbate–adsorbent interaction at constant temperature [21]. The following is the linearized form of the Langmuir Equation (3):

$$\frac{1}{qe} = \frac{1}{KL q_{max}} \times \frac{1}{Ce} + \frac{1}{q_{max}} \quad (3)$$

where KL and q_{max} are the Langmuir constants for adsorption energy and maximum adsorption capacity, respectively. Ce and qe denote the equilibrium concentration (mg/L) and amount adsorbed at equilibrium time (mg/g), respectively [22]. The Langmuir isotherm model may be used to explain adsorption equilibrium conditions and provide parameters (q_{max} and KL) for quantitatively evaluating adsorption behaviors in various adsorbate–adsorbent systems [23]. The values of KL and q_{max} may be computed by graphing $1/(qe)$ against $1/(Ce)$ [24]. The Langmuir adsorption isotherm can alternatively be represented by the equilibrium parameter, RL , a dimensionless constant separation factor, as shown in Equation (4) below:

$$RL = \frac{1}{1 + KL \times C_i} \quad (4)$$

where KL is the Langmuir constant, and C_i is the initial concentration in mg/L.

Meanwhile, the empirical isotherm of Freundlich is a suitable isotherm for adsorption from solution, being the most suitable isotherm for describing π - π interaction, H-bonding,

and electrostatic attraction [25]. The logarithmic form can be conveniently written as Equation (5):

$$\ln q_e = \ln K_f + \frac{1}{n} \times \ln C_e \quad (5)$$

where K_f and n stand for the adsorption capacity and intensity, respectively. The intercept and slope of the plot of $\ln q_e$ vs. $\ln C_e$ may be used to obtain the constants K_f and n .

2.5. Municipal Wastewater Samples

Wastewater samples were collected from three local drains (Drainages) in the city of Ramadi (Table 1). These drains were selected from a densely populated area where nearby houses discharge large quantities of untreated wastewater directly into the Euphrates River.

Table 1. Sampling sites of Ramadi City drainages.

Site Number	GPS	
	X	Y
S1	33.464930	43.245072
S2	33.463824	43.340273
S3	33.435416	43.266315

This wastewater carries a high concentration of various pollutants that flow directly into the Euphrates River without any prior treatment. Wastewater samples were collected from the center and sides of the waterway using 500 mL pre-sterilized, dark-colored glass containers with tight-fitting lids. The collected samples were taken to the laboratory for chemical analysis, including the measurement of heavy metal concentrations under optimal biosorption conditions.

2.6. Immobilization Technique

This technique was used to fix or trap the plant wastes of PHP into alginate solution or porous polymer to form beads. The PHP biomass was immobilized by entrapment in the polymer matrix of Na-alginate (Fluka, Buchs-Switzerland), solving 2 g of Na-alginate in 100 mL of distilled water (2% *w/v*), and then placed on a magnetic stirrer for well mixing. After that, peanut powder (biomass) was added to the sodium alginate solution at a specific weight of 5 g/L. However, the alginate biomass slurry was introduced into 0.1 M of CaCl_2 (Fluka, Buchs-Switzerland) (dissolving 5.549 g of CaCl_2 in 500 mL of deionized distilled water DDW) for polymerization and bead formation using a 5 mL syringe. The resultant beads were about 4 mm in diameter, 0.9 g in weight, and exhibit excellent mechanical stability, allowing them to be used in adsorption processes without breaking down or leaching the peanut hulls into the surrounding environment. However, the beads are highly sensitive to pH levels, shrinking in acidic solutions and expanding in alkaline solutions (Figure 2). The hull-entrapped beads were cured in CaCl_2 solution for 30 min., washed twice with 200 mL of DDW, and then preserved in 5 mM of CaCl_2 (dissolving 0.278 g of CaCl_2 in 500 mL of DDW) at 4 °C for further use [26].

2.7. Laboratorial Design of Treatment Unit

The laboratory system was designed for biological treatment depending on the continuous flow system technique, using an acrylic column containing the biomass beads attached to PHP. Generally, the treatment unit includes three columns: the first one was employed to receive the polluted water doses, and the last one was created to collect the treated water. However, the most important column is the middle one, which holds the biosorption process containing the biomass beads responsible for the treatment and

decontamination of polluted samples. This column has a diameter of 5.6 cm and a height of 41 cm, occupies a volume of about 1009 mL as a reaction area, and receives water from a water pump through a flow-meter at a velocity of 2 L/h equipped with different filters and valves to control the flow in room temperature, as seen in Figure 3. However, the resulting water was gathered from the last column for heavy metal concentration measurements by FAAS.

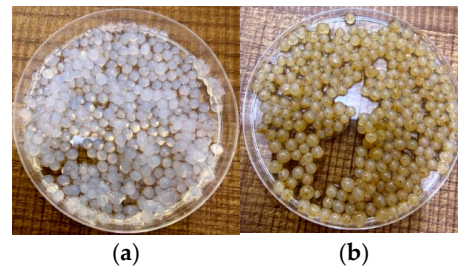


Figure 2. Free and immobilized beads: (a) Free Ca-alginate without PHP; (b) PHP immobilized to Ca-alginate.

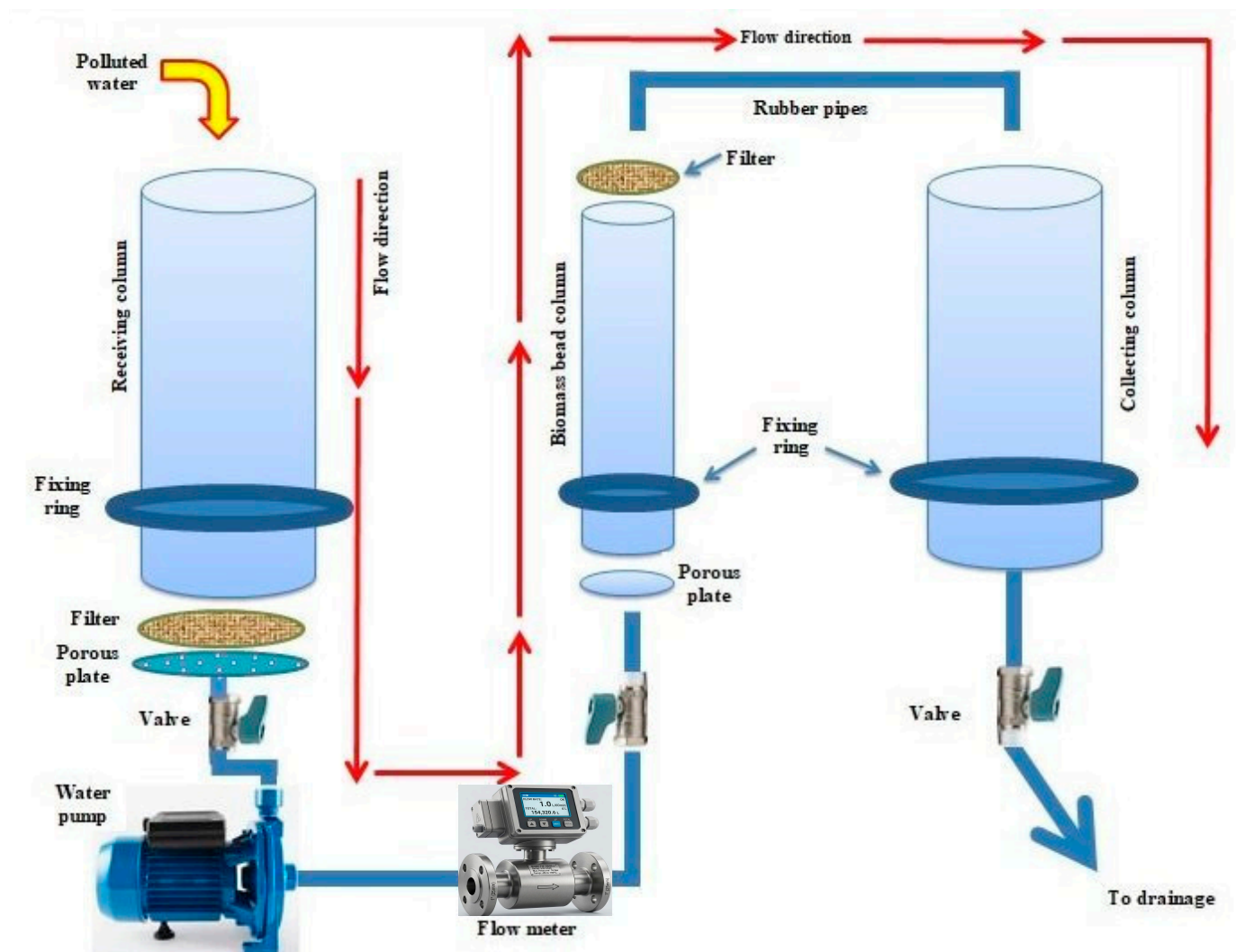


Figure 3. Laboratory treatment unit diagram.

2.8. Statistical Analysis

A one-way analysis of variance (ANOVA) was employed to evaluate the significance of differences among the experimental data obtained under various operating conditions.

Statistical analyses were performed using IBM SPSS Statistics (2022 version). In addition, all nonlinear regression analyses, parameter estimations, and graphical representations of the Langmuir and Freundlich isotherm models were carried out using Origin Lab-Origin software (Version 8). The level of statistical significance was considered to ensure the reliability and reproducibility of the obtained adsorption results.

3. Results and Discussion

In order to determine the ideal conditions for such variables to achieve the highest removal of heavy metals as hazardous environmental pollutants, the current study examines the effects of several environmental parameters, including pH, temperature, powder size and weight, and retention time, on the efficiency of the biosorption process using *Arachis hypogaea* hull biomass for the removal or reduction in heavy metal ions at a constant and uniform primary concentration of 100 mg/L.

3.1. *Arachis hypogaea* PH and PHP Biosorption

Both *Arachis hypogaea* PH and PHP have the potential to perform biosorption for pollutant removal, but their efficiency and mechanism differ due to differences in composition and structure [27]. The pH value of 7 was kept throughout the experiment to ensure clear and purely negative charges on the adsorbent surfaces, which contributes to an increased affinity for positively charged metal ions via electrostatic interaction [17]. Because the pH of 7 is a critical value, heavy metal ions are precipitated when the pH value exceeds 7, causing interference between the adsorption and precipitation processes [21]. Preliminary results indicated an overall increase in the efficiency of the biosorption process when using PHP compared to PH. Statistical analysis of the results showed significant differences ($p < 0.001$) in the efficiency of biosorption for both biomass forms of the studied heavy metals. Also, LSD = 2.378 at $p > 0.05$ showed that this variance in the results indicate that PHP outperformed PH in removing heavy metal ions, which enhances the use of these biomass, especially since they can be obtained ready-made and almost without financial costs through local confectionery, chocolate, and peanut butter factories that produce such waste. PHP removal average values (84.63 ± 4.6 , 77.14 ± 3.7 , 74.85 ± 2.3 , $65.91 \pm 1.9\%$) for Pb, Zn, Cu, and Cd, respectively, were found to be higher than those observed for PH, which recorded (Pb = 61.13 ± 1.6 , Zn = 49.44 ± 1.1 , Cu = 34.15 ± 0.9 , and Cd = $26.33 \pm 0.9\%$), as shown in Figure 4.

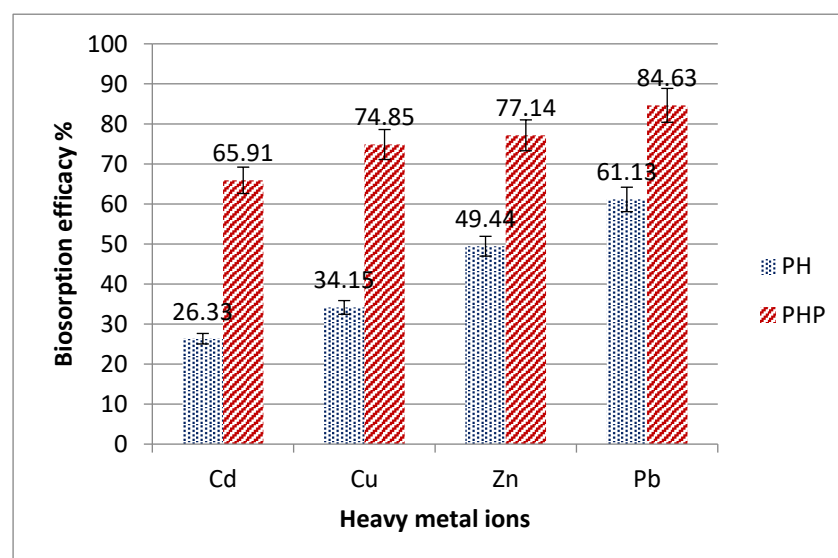


Figure 4. Comparison of the efficiency of the biosorption process in peanut hulls and hull powder.

The biosorption efficiency using PHP indicates different binding affinities for the studied metals. However, to increase the efficiency of the biosorption process, PHs may need some modifications by activating with sulfuric acid, which increases the formation of effective functional groups of $-\text{SO}_3\text{H}$, and other modifications that enhance the efficiency of the treatment process [28]. Meanwhile, PHP consists of a high percentage of cellulosic fibers, carbohydrates, and various proteins; all of these materials are rich in oxygen-containing functional groups like carboxyl and hydroxyl groups that can bind heavy metal ions easily, quickly, and effectively through the electrostatic attraction mechanism [29]. Therefore, PHP does not require any modifications that may be economically unfeasible, as it can be used directly in the treatment process. Thus, PHP was chosen for subsequent biosorption experiments in this study.

3.2. Point of Zero Charge Determination (pH_{pzc})

The point of zero charge is a key parameter in biosorption studies, providing insights into the surface charge characteristics of a material and its adsorption behavior. In this study, the pH_{pzc} of peanut hull powder biomass was determined to be 6.2 (Figure 5), indicating a negatively charged surface at pH levels above these values and a positive charge below these points.

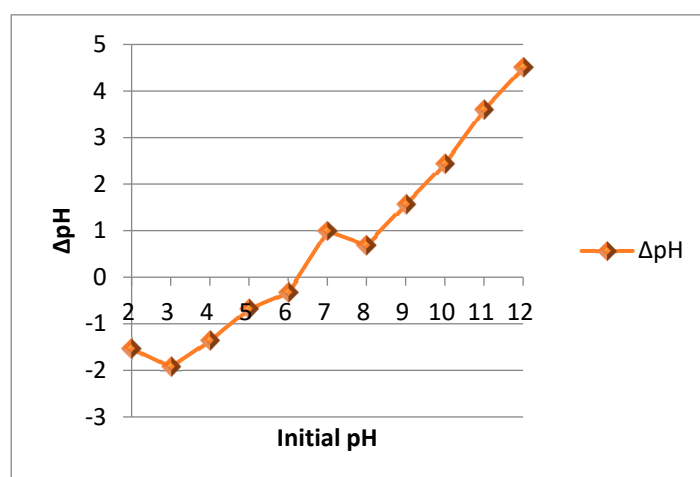


Figure 5. Point of zero charge (pH_{pzc}) curve of PHP at 30 °C, an agitation speed of 120 rpm, a biosorbent dose of 5 g/L, and a pH range of 2–12.

This finding is crucial, as it identifies the optimal pH range for heavy metal adsorption, which is dependent on surface charge. Compared with other plant-based biosorbents, the used plant biomass in this study exhibits a pH_{pzc} similar to that of *Moringa oleifera* bark, which was reported as 6.0 [30], while Yohimbe bark shows a slightly higher pH_{pzc} of 7.1 [31]. These findings underscore the importance of pH_{pzc} as a key parameter in evaluating and optimizing biosorbent performance.

3.3. Biosorption Characteristics and Optimization

For this experiment, different values for pH, temperature, retention time, adsorbent weight, and particle size were selected to determine the optimal conditions for biosorption while fixing the other parameters. The results indicated that the optimal conditions were pH = 6, 40 °C, 60 min., 0.5 g, and 0.5 mm, respectively, as follows:

3.3.1. Effect of pH

The solution pH was found to be a critical controlling factor for the biosorption performance of *Arachis hypogaea* biomass towards the uptake of heavy metal ions. The

results clearly indicated that pH significantly affected the adsorption efficiency of all the metals studied (Pb, Zn, Cu and Cd). Overall, the highest biosorption efficiencies were obtained at pH 6. The maximum removal percentage was for Pb ($96.54 \pm 6.93\%$), followed by Zn ($95.72 \pm 7.84\%$), Cu ($94.98 \pm 7.08\%$), and Cd ($93.17 \pm 7.04\%$) (Figure 6).

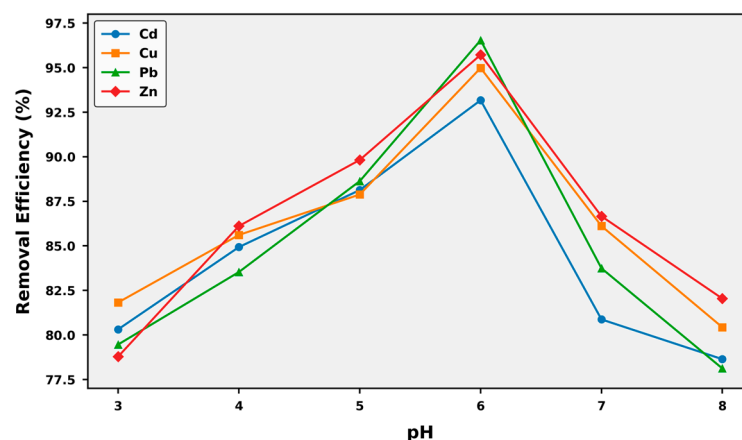


Figure 6. Effect of solution pH on removal efficiency of peanut hull powder (at constant conditions of time, 60 min., temp. 30 °C, dose 10 g/L).

The biosorption efficiency of PHP biomass was relatively low for all metals under strongly acidic conditions (pH 3–5) from $78.78 \pm 5.88\%$ for Zn at pH 3 to $88.63 \pm 5.87\%$ and $89.82 \pm 6.28\%$ for Pb and Zn, respectively, at pH 5. This reduction could be due to the high competition between the hydrogen ions (H^+) and metal cations for the available active binding sites on the surface of the biosorbent [32], in addition to the synergistic effect of functional groups on the biosorbent surface, which enhance the electrostatic attraction between the biosorbent and metal ions [33]. However, the biosorption efficiency was slightly decreased above pH 6 with values ranging between $78.12 \pm 5.66\%$ for Pb at pH 8 and $86.66 \pm 6.76\%$ for Zn at pH 7. This reduction might be due to the possible formation of the metal hydroxide complexes and the reduced availability of free metal ions in the solution [34]. The present results are in good agreement with some recent studies that have pointed out solution pH as a critical controlling parameter in the biosorption of heavy metals using plant-based biosorbents including *Arachis hypogaea*. The optimum removal efficiency at near-neutral pH (pH = 6) is in agreement with the work of Caglar and Kayranli [35], who stated that peanut shell-based biosorbents exhibited adsorption performance under neutral conditions due to enhanced deprotonating of surface functional groups such as hydroxyl and carboxyl groups, leading to a higher density of negatively charged active sites [36].

The increase in the pH of the solution improves the adsorption efficiency because of the increasing electrostatic attraction between positively charged metal ions and the negatively charged surface of the biosorbent. Moreover, the observed trend in metal removal efficiency ($Pb > Zn > Cu > Cd$) agreed with the results reported by Anggayasti et al. [37], who attributed the variations to the difference in the physicochemical properties of the metal ions, particularly their hydrated ionic and charge densities, which control their affinity to the available binding sites on the biosorbent surface [38]. However, slight differences were observed when the present results were compared with some studies in which the optimum biosorption performance of *Arachis hypogaea* was observed at slightly higher pH values (around 7) [39]. This might be attributed to the differences in the biosorbent preparation methods, particle size distribution, and surface activation procedures, which can greatly affect the ionization characteristics of functional groups and the accessibility of active adsorption sites on the biosorbent surface [40]. Yet, statistical analysis using

one-way ANOVA demonstrated that the effect of pH on biosorption efficiency was highly significant ($p \leq 0.01$) for all investigated metals. In addition, significant differences were identified among the biosorption efficiencies of the studied metals, as confirmed by the least significant difference (LSD) value of 2.329%, indicating pronounced variability in the affinity of metal ions toward the biosorbent under different pH conditions. Overall, the findings of this study highlight that pH is the primary controlling parameter governing the biosorption process using *Arachis hypogaea* PHP biomass. This is because pH directly influences surface charge development, metal ion speciation in solution, and the strength of electrostatic interactions and complexation mechanisms responsible for the removal of heavy metal ions from aqueous media.

3.3.2. Effect of Contact Time

Contact time is a key operational parameter that significantly affects the biosorption efficiency of heavy metal ions onto *Arachis hypogaea* PHP biomass, as it directly regulates the duration of interaction between metal ions and the available active binding sites on the biosorbent surface. The results demonstrated a substantial increase in biosorption efficiency with increasing contact time until the attainment of equilibrium conditions (Figure 7).

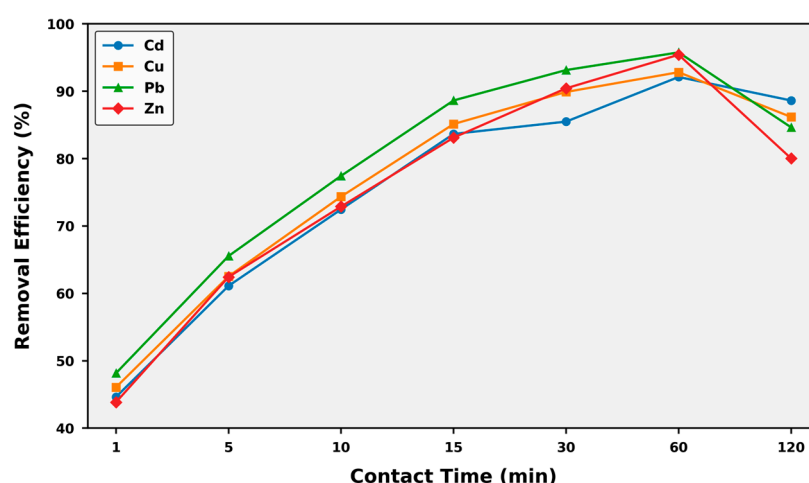


Figure 7. Effect of contact time on removal efficiency of peanut hull powder (under constant conditions of pH 6, temp. 30 °C, dose 10 g/L).

At relatively short contact times, particularly during the initial stages of adsorption, low biosorption efficiencies were observed for all investigated metals. The recorded values ranged at $43.85 \pm 4.93\%$ for Zn followed by values of 44.63 ± 5.31 , 46.07 ± 4.97 , and $48.16 \pm 5.02\%$ for Cd, Cu and Pb, respectively, within the first minute of the process. The progressive increase in adsorption efficiency with time can be attributed to the progressive occupation of the abundant available active sites on the surface of PHP biomass [41]. During the initial stages of adsorption, a high concentration gradient exists between the solution and the biosorbent, which enhances the mass transfer of metal ions toward the surface and facilitates their attachment to the active binding sites [42,43]. However, contact time was found to be a significant parameter in the biosorption process and the removal efficiency of all heavy metals under study, which increased until equilibrium was attained at 60 min. This optimum contact time showed the maximum biosorption efficiency for Pb ($95.76 \pm 7.08\%$), followed by Zn ($95.41 \pm 6.79\%$), Cu ($92.82 \pm 6.64\%$) and Cd ($92.11 \pm 7.33\%$). The enhanced metal uptake can be attributed to the presence of active sites for adsorption and high affinity of metal ions towards oxygen-bearing functional groups such as hydroxyl and carboxyl groups naturally existing in PHP biomass. The presence of such groups helps

in the adsorption process through ion exchange and complexation reactions, leading to increased biosorption efficiency [44].

The results showed that the biosorption performance of PHP was negatively affected by increasing the contact time beyond 60 min. The adsorption increased with the increase in contact time, but the removal efficiency was significantly decreased at 120 min for all studied metals. This behavior can be explained by the progressive saturation of the active sites and the reaching of a dynamic equilibrium between the adsorbed and dissolved metal ions [21]. Under prolonged contact conditions, the desorption processes may become more important and lead to a reduction in the net metal uptake. These observations are in agreement with previous reports, including Rambabu et al. [45], who attributed the decrease in adsorption efficiency at longer contact time to site saturation and the effects of the adsorption–desorption equilibrium. Adsorption using *Phoenix dactylifera*-based biosorbents reaches equilibrium rapidly, followed by a decline or stabilization due to occupation of active sites [45]. Additionally, these findings further support the hypothesis that excessive contact times may adversely affect adsorption performance due to the occurrence of desorption processes and the redistribution of metal ions between the solid biosorbent and the surrounding solution. The establishment of adsorption–desorption equilibrium under prolonged agitation conditions can reduce the net uptake of metal ions, leading to a decline in overall removal efficiency. However, this behavior is not universally observed across all biosorbent systems. Several studies have reported either a continuous increase or a stabilization of adsorption efficiency at extended contact times without a significant decline. Such variations have been attributed to stronger metal–biosorbent interactions, enhanced surface affinity, and lower desorption rates, which are largely influenced by the physicochemical properties of the biosorbent, including surface activation, functional group availability, and pore structure characteristics [46,47].

Statistical evaluation using one-way analysis of variance (ANOVA) revealed that contact time exerted a highly significant influence ($p \leq 0.01$) on the biosorption efficiencies of all investigated heavy metal ions. The presence of significant differences among the mean adsorption values was further confirmed by the least significant difference (LSD) value of 1.254, highlighting the critical role of contact time in governing the adsorption process. These results demonstrate that biosorption performance is strongly dependent on the attainment of equilibrium conditions and the duration of interaction between metal ions and the biosorbent surface. Therefore, optimization of contact time is essential for maximizing the biosorption capacity of *Arachis hypogaea* biomass and enhancing its effectiveness in the removal of heavy metals from aqueous systems.

3.3.3. Effect of Temperature

Temperature is an important environmental parameter that strongly influences the PHP biosorption performance of *Arachis hypogaea* biomass, as it controls the mobility of metal ions, the diffusion rates and the reactivity of functional groups present on the surface of the biosorbent. Figure 8 depicts the effect of solution temperature on the removal efficiencies of Cd, Cu, Zn and Pb ions.

The obtained results revealed that the biosorption efficiency gradually increased with the increase of temperature up to 40 °C, where the maximum removal efficiencies for all investigated heavy metals were recorded: Pb ($95.2 \pm 6.94\%$) followed by 92.66 ± 8.12 , 92.24 ± 6.72 , and $91.86 \pm 7.04\%$ for Zn, Cu, and Cd, respectively.

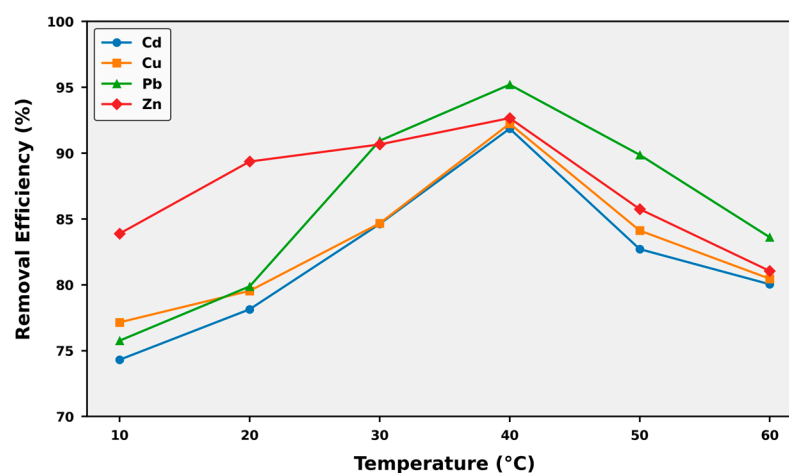


Figure 8. Effect of temperature on removal efficiency of peanut hull powder (under constant conditions of pH 6, time 60 min., dose 10 g/L).

The enhanced adsorption performance at moderate temperature can be explained by the increased kinetic energy of metal ions, which favors their diffusion from the bulk solution to the active binding sites on the PHP surface. In addition, mild heating may activate the surface and strengthen the interactions between metal ions and oxygen-containing functional groups, especially hydroxyl and carboxyl groups, thereby increasing the accessibility and reactivity of adsorption sites [48]. The biosorption efficiency significantly reduced at lower temperatures especially at 10 °C with removal efficiencies of $74.31 \pm 5.93\%$ for Cd to $83.89 \pm 6.11\%$ for Zn. This reduction can be ascribed to the limited mobility of metal ions and the lower diffusion rates at low temperatures, which restrict effective interactions with the biosorbent surface [49]. However, when the temperature exceeded 40 °C, a marked decline in biosorption efficiency was observed. At 60 °C, the removal efficiencies decreased, ranging from $80.04 \pm 6.06\%$ for Cd to $83.61 \pm 5.49\%$ for Pb. This reduction may be attributed to the partial disruption of weak intermolecular interactions between metal ions and the biosorbent surface, along with possible structural modifications of surface functional groups under elevated temperature conditions [50]. When the temperature gets too high, the active binding regions on the biosorbent change their form and characteristics, which modifies how well they can trap metals. However, the specific structural changes that occur to biosorbent surface groups at high heat may include the thermal degradation which disrupt the chemical bonds of active groups such as carboxyl, and amine. Functional group conversion as heat can change one group into another. Dehydration results in the shrinkage or cross-linking of carbohydrate chains in materials like cellulose, which eliminates binding sites. Protein denaturation disrupts the particular structural forms required to capture metal ions effectively [48–50]. In general, temperatures above 60 °C usually decrease biosorption capacity because the structural degradation prevents heavy metals from adhering correctly [46].

These results suggest that biosorption using PHP biomass is most effective under moderate thermal conditions. Thus, the present study demonstrates that optimum biosorption performance is achieved at intermediate temperatures, whereas higher temperatures lead to a decline in adsorption efficiency, likely due to the weakening of surface interactions and the destabilization of adsorbate–adsorbent binding forces. Additionally, statistical analysis revealed that solution temperature had a highly significant effect ($p \leq 0.01$) on the biosorption efficiency of all investigated heavy metals. Moreover, the least significant difference (LSD = 2.237%) confirmed the presence of statistically significant differences among the adsorption percentages, highlighting the crucial role of temperature in govern-

ing the adsorption capacity and performance stability of PHP biomass during heavy metal removal from aqueous solutions.

3.3.4. Effect of Biosorbent Weight

The biosorbent dosage is a key operational parameter that significantly influences the efficiency of heavy metal removal using PHP biomass, as it directly governs the availability of active adsorption sites and the overall surface area accessible for metal ion binding. The effect of biosorbent dosage on the removal efficiencies of Cd, Cu, Zn, and Pb ions is presented in Figure 9.

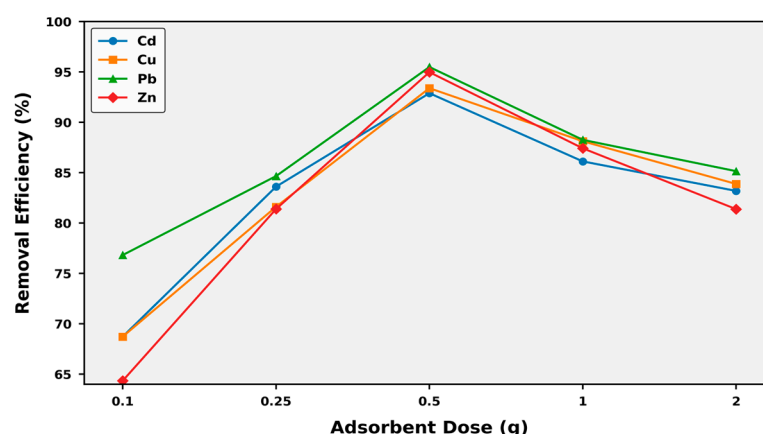


Figure 9. Effect of adsorbent dose on removal efficiency of peanut hull powder (under constant conditions of pH 6, temp. 40 °C, time 60 min).

The experimental results demonstrated that a biosorbent dosage of 0.5 g achieved the highest biosorption efficiency for all investigated heavy metals. Among the tested metals, Pb exhibited the highest removal efficiency ($95.49 \pm 7.35\%$), followed by Zn and Cu with values of $94.98 \pm 7.14\%$ and $93.39 \pm 6.73\%$, respectively, while Cd recorded the lowest mean removal efficiency of ($92.89 \pm 7.03\%$). The enhanced performance observed at a biosorbent dosage of 0.5 g can be attributed to the optimal balance between the number of available active binding sites and the effective dispersion of PHP particles within the solution, which facilitates greater accessibility of metal ions and improves surface interactions. In contrast, lower biosorbent dosages (0.1 and 0.25 g) resulted in significantly reduced biosorption efficiencies for all investigated metals. This decrease may be attributed to an insufficient number of available adsorption sites to effectively accommodate the metal ions present in the solution, resulting in incomplete removal [51]. Conversely, increasing the biosorbent dosage beyond the optimum level (1 and 2 g) led to a noticeable reduction in adsorption efficiency. This decline may be attributed to particle agglomeration and the overlapping of adsorption sites at higher biomass concentrations, which can decrease the effective surface area available for adsorption and hinder the accessibility of active binding sites. As a result, the interaction between metal ions and the functional groups responsible for adsorption becomes less efficient [52].

The adsorption behavior observed in the present study underscores the critical role of the heterogeneous surface characteristics of PHP biomass and its oxygen-containing functional groups in governing metal ion uptake. These functional groups serve as the primary binding sites for metal adsorption through mechanisms such as ion exchange and/or surface complexation. Therefore, the optimization of biosorbent dosage is essential to achieve maximum adsorption efficiency while ensuring the effective utilization of the biosorbent material and minimizing unnecessary biomass consumption [53]. One-way ANOVA analysis revealed that biosorbent dosage exerted a highly significant effect

($p \leq 0.01$) on the removal efficiencies of all investigated heavy metals. In addition, the least significant difference (LSD) value of 1.503 confirmed the presence of statistically significant differences among the mean adsorption values.

3.3.5. Effect of Particle Size

Particle size is considered a significant physical parameter affecting the biosorption efficiency of PHP, as it directly influences the available surface area, accessibility of active adsorption sites, and diffusion behavior of metal ions toward the biosorbent surface. The mean biosorption percentages of Cd, Cu, Zn, and Pb adsorbed by PHP at different particle sizes are presented in Figure 10.

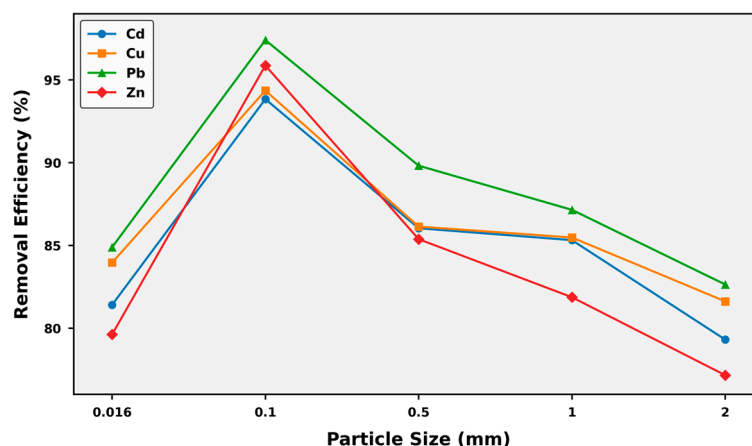


Figure 10. Effect of particle size on removal efficiency of peanut hull powder (under constant conditions of pH 6, temp. 40 °C, time 60 min. dose 5 g/L).

The particle size of 0.1 mm exhibited the highest average biosorption efficiencies for all investigated heavy metals. The corresponding removal percentages were $97.41 \pm 7.33\%$ for Pb, $95.87 \pm 6.05\%$ for Zn, $94.36 \pm 6.78\%$ for Cu, and $93.84 \pm 7.06\%$ for Cd. The enhanced adsorption performance observed at this intermediate particle size can be attributed to an optimal balance between available surface area and particle stability. This balance increases the accessibility of active surface sites and improves the diffusion and transport of metal ions within the biosorbent, thereby promoting more efficient binding and adsorption processes [54]. However, at the smallest particle size (0.016 mm), the lowest biosorption efficiencies were recorded for all investigated metals. Although finer particles typically offer a higher specific surface area, excessively small particle sizes may promote agglomeration and restrict pore accessibility, thereby reducing the effective contact between metal ions and available adsorption sites. When the particle size increased, a notable improvement in biosorption efficiency was observed, which can be attributed to better particle dispersion and improved exposure of active surface functional groups. Nonetheless, a further increase in particle size to 1 and 2 mm led to a gradual decrease in adsorption efficiency, likely due to the reduced specific surface area and the limited availability of accessible binding sites. Meanwhile, at larger particle sizes of approximately 2 mm, the biosorption efficiencies decreased to 82.64 ± 5.42 , 77.16 ± 5.08 , 81.62 ± 5.06 , and $79.31 \pm 5.11\%$ for Pb, Zn, Cu, and Cd, respectively. This decline can be attributed to the reduced specific surface area and the limited diffusion of metal ions into the internal active adsorption sites within larger particles, which consequently restricts the overall biosorption capacity [55].

The findings of the present study are in agreement with those reported by Elkholy et al. [56], who demonstrated that the adsorption of heavy metals is highly dependent on the surface characteristics of biosorbents. Statistical analysis revealed that particle size exerts a

highly significant influence ($p \leq 0.01$) on the biosorption efficiencies of all examined heavy metals. In addition, the least significant difference (LSD = 0.961) confirmed statistically significant variations among the mean adsorption values, indicating that particle size is a critical parameter governing the adsorption performance of *Arachis hypogaea* hull powder (PHP).

3.4. FTIR Analysis

Figure 11 of PHP FTIR spectra (under optimal conditions) shows broad O-H and N-H stretch at (3400 cm^{-1}) [57], aliphatic C-H stretch at $2920\text{--}2850\text{ cm}^{-1}$ [58], C=O for amide I at $1650\text{--}1600\text{ cm}^{-1}$ [59], Amide II for N-H bend and C-N at $1550\text{--}1500\text{ cm}^{-1}$ [60], COO^- and CH bend at $1400\text{--}1300\text{ cm}^{-1}$ [61], and C-O for polysaccharide at $1200\text{--}1000\text{ cm}^{-1}$ [62]. Metal adsorption causes the wide O-H and N-H stretching band (3424 cm^{-1}) to move to higher wavenumbers, indicating weakening of hydrogen bonds and coordination with hydroxyl or amine groups. The C-H stretching vibrations (2922 cm^{-1}) are almost unchanged, indicating that aliphatic groups are not implicated. The carbonyl (C=O) and amide I regions (1642 cm^{-1}) exhibit negligible changes, but amide II (1542 cm^{-1}) moves to lower values, indicating partial coordination through nitrogen atoms of amide links. Nevertheless, the carboxylate band (1418 cm^{-1}) changes to higher frequencies, indicating metal binding to COO^- groups. Metal adsorption resulted in new absorption bands in the low-frequency FTIR spectra ($400\text{--}700\text{ cm}^{-1}$), indicating the creation of metal–oxygen (M–O) bonds.

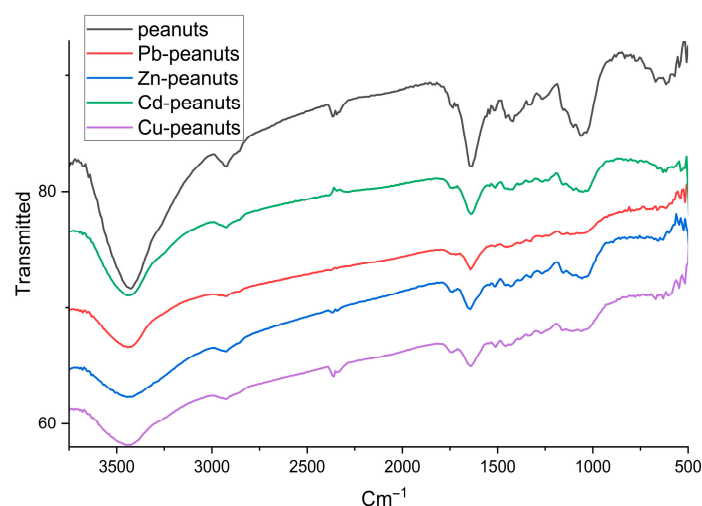


Figure 11. FTIR analysis for PHP before and after biosorption.

The Zn-loaded peanut sample exhibited a medium-intensity band about 600 cm^{-1} , indicating the stretching vibration of the Zn–O bond [63]. The Cd-loaded sample displayed a faint, wide band about 590 cm^{-1} , indicating Cd–O vibration [64]. The Pb-loaded spectra showed a clear band about 520 cm^{-1} , confirming the presence of Pb–O stretching [65]. The Cu-loaded sample showed a stronger absorption at 580 cm^{-1} , indicating the Cu–O stretching mode [66]. However, FTIR research indicates that adsorption of Zn^{2+} , Cd^{2+} , Pb^{2+} , and Cu^{2+} ions occurs largely through oxygen- and nitrogen-containing functional groups on the surface of peanut powder.

3.5. SEM Biomass Investigation

SEM (Model: Tescan–Vega III, Czech Republic) was used to examine the morphology of raw biomass and one that is metal-treated under optimal conditions. A mixed solution with 100 ppm of each of the heavy metals under study—copper, cadmium, lead, and zinc—was created. This solution was then combined with one gram of PHP biomass, and

the mixture was allowed to settle for two hours before a sample was obtained for scanning electron microscopy (SEM) analysis.

Yet, the morphology of the heavy metal mixture treated with PHP biomass and the untreated mixture was examined. Figure 12A displays PHP biomass prior to biosorption at magnifications of 1.00 kx and 5.00 kx, while Figure 12B displays the biomass following biosorption of the heavy metals under study at magnifications of 1.00 kx and 5.00 kx, respectively. However, a flat, dark-colored layer with a few small, light-colored patches with slightly scaly structures is present in the primal inquiry of Figure 12A—1.00 kx. These areas comprise simple, sparsely distributed masses with smooth, soft surfaces that are around 10 microns in size (Figure 12A—5.00 kx). Meanwhile, Figure 12B—1.00 kx once more explains the existence of flat, overlapping layers that are typically light in color, have distinctly shiny areas of higher crustal content, and have a homogeneous mass density distribution with surface roughness structured in multiple fiber-like crustal layers that contain particles of 10 microns in size (Figure 12B—5.00 kx).

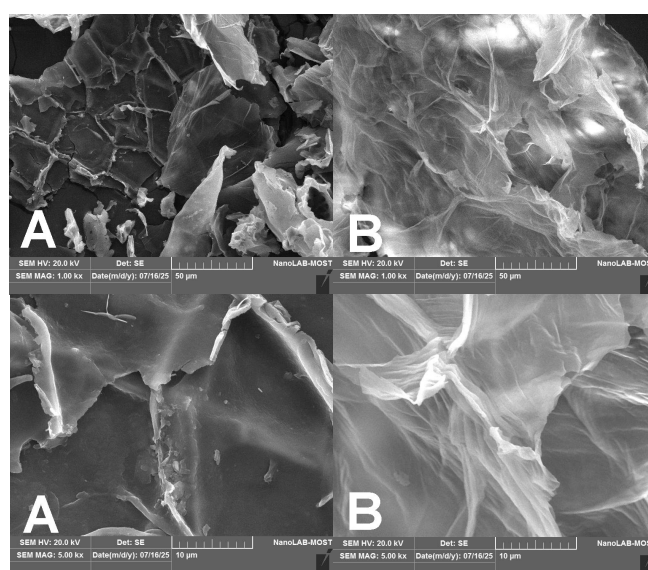


Figure 12. SEM images of (A) peanut hull biomass before biosorption; (B) peanut hull biomass after Pb^{2+} , Zn^{2+} , Cu^{2+} , and Cd^{2+} biosorption at 1.00 kx and 5.00 kx.

However, the dark zones include organic proteins and cellulose of carbon-rich fibers. Meanwhile, the changing-intensity light-colored portions contain compounds of inorganic elements like sodium or phosphorus [67]. Additionally, the precipitation of heavy metal ions onto the surface of the dried biomass was responsible for the observed changes in biomass shape [68]. When Cd was eliminated from the aqueous solution, *Parachlorella* sp. showed similar outcomes [69]. The cross-linking of functional groups and metal ions may be responsible for the biomass's morphological alterations [70,71].

3.6. Adsorption Isotherms

3.6.1. Langmuir Isotherm

The Langmuir isotherm model may be used to explain adsorption equilibrium conditions and provide parameters (q_{max} and KL) for quantitatively evaluating adsorption behaviors in various adsorbate–adsorbent systems [72]. The values of KL and q_{max} may be computed by graphing $1/(qe)$ against $1/(Ce)$ [24]. The correlation coefficients R^2 for the adsorption of all metal ions were more than 0.91, as shown in Figure 13.

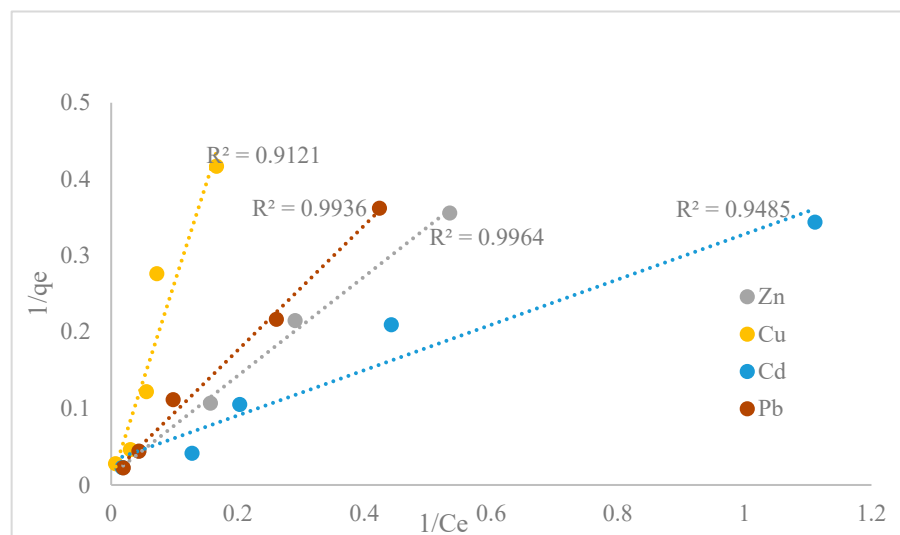


Figure 13. Langmuir isotherm for removing metal ions using PHP.

As indicated in Table 2, the amount of metal ions needed to form a monolayer on the substrate's surface (q_{max} in mg/g) was computed from (1/intercept), and the constant affinity of the adsorption sites (KL in L/mg) was computed from 1/(slope q_{max}). Nevertheless, from the data in Table 2, the q_{max} values found to be varied with the charge intensity and the ionic radius. Therefore the sequences of q_{max} for the ions must be arranged as follows: $Cu^{2+} > Zn^{2+} > Cd^{2+} > Pb^{2+}$. But it is obvious the q_{max} of Cd^{2+} is less than Pb^{2+} , which may be attributed to the fact that the cadmium (Cd^{2+}) has the greatest Langmuir constant (KL), indicating considerable affinity for adsorption sites. However, it has the lowest adsorption capacity (q_{max}) of the investigated ions. Cd^{2+} binds robustly, but only at specific high-energy sites. Copper (Cu^{2+}) has the highest adsorption capacity, indicating a large number of accessible sites for adsorption, despite a lower affinity per site (KL) [73].

Table 2. The values of q_{max} and KL for the metal ions with PHP adsorbents.

Peanut Hull Powder PHP			
Metal	q_{max}	KL	R^2
Cd^{2+}	31.66	0.083	0.94
Cu^{2+}	178.6098	0.013	0.91
Pb^{2+}	77.90592	0.015	0.99
Zn^{2+}	78.16026	0.0046	0.99

The Langmuir adsorption isotherm can alternatively be represented by the equilibrium parameter, RL [72,73]. Table 3 displays the RL values that were computed for each concentration and metal ion.

Table 3. The RL values at various concentrations and different metal ions.

Peanut Hull Powder PHP				
Conc.	Cd	Cu	Pb	Zn
30	0.285	0.709	0.679	0.878
50	0.193	0.594	0.559	0.812
100	0.107	0.422	0.388	0.684
250	0.046	0.226	0.203	0.464
500	0.023	0.128	0.113	0.302

According to Table 4, the parameter RL indicates the type of isotherm shape. As long as $0 < RL < 1$, this kind of Langmuir isotherm is favorable for all metal ions and C_i concentrations, according to Table 4's interpretation of RL values [74]. Consequently, all of the experimental data are fitted using the Langmuir isotherm since the RL is favorable and the correlation coefficient is more than 0.95, as seen in Figure 11.

Table 4. The interpretation of RL values.

Type of Isotherm	RL Value
unfavorable	$RL = 1$
linear	$RL > 1$
favorable	$0 < RL < 1$
irreversible	$RL = 0$

3.6.2. Freundlich Isotherm

The empirical isotherm of Freundlich is a suitable isotherm for adsorption from solution. The most suitable isotherm for describing interaction, H-bonding, and electrostatic attraction is the Freundlich isotherm [25].

However, with an R^2 of more than 0.88, the link between $\ln q_e$ and $\ln C_e$ in Figure 14 was shown to be strongly connected. Table 5 displays the evaluated constants. Hence, the fact that every metal ion adsorption had n values greater than one, as shown in Table 5, suggests that the adsorption process is physical and highly heterogeneous [23–75]. Large adsorption capacity (high K_F value) was not correlated with strong affinity (high n value). Particle characteristics including size distribution, specific surface area, and surface functional groups were used to calculate the K_F . As seen in Figure 12, the adsorption isotherms for every metal ion were fitted to the Freundlich n with a correlation coefficient more than 0.88 and n values greater than 1 [23–76].

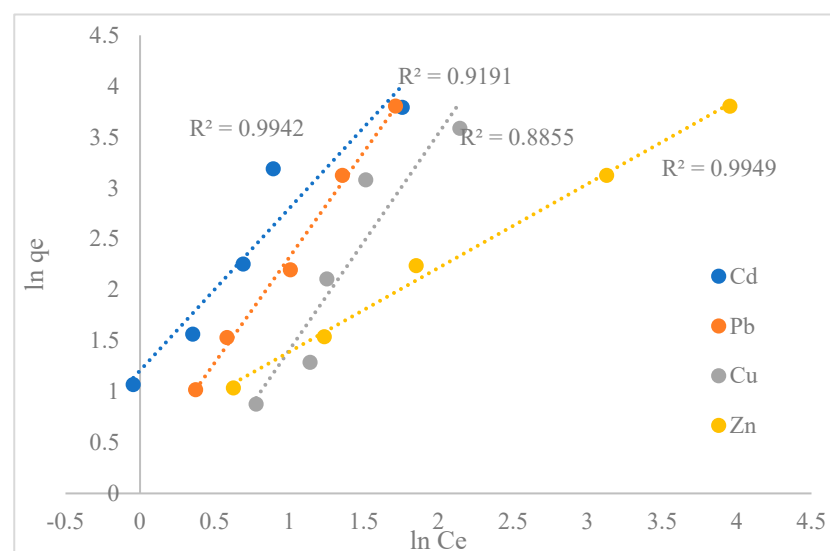


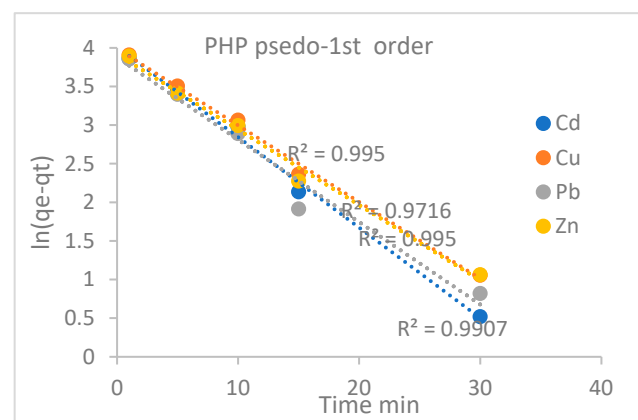
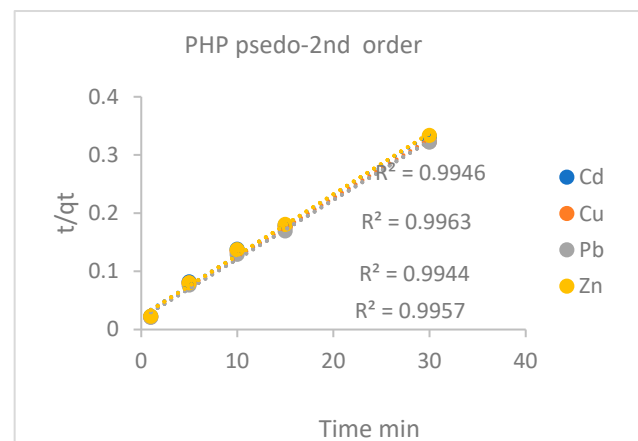
Figure 14. Freundlich isotherm for removing metal ions using PHP.

Table 5. The values of n and K_f for the metal ions with the adsorbent.

Peanut Hull Powder PHP			
Metal	K_f	n	R^2
Cd^{2+}	2.84	1.33	0.91
Cu^{2+}	1.61	1.92	0.88
Pb^{2+}	2.11	1.35	0.99
Zn^{2+}	5.40	17.81	0.94

3.7. Biosorption Kinetics

Based on the kinetic parameters and correlation coefficients from Figures 15 and 16 and Table 6, it can be noted that pseudo-second-order model showed outstanding correlation coefficients R^2 of 0.99. Additionally, the computed equilibrium adsorption capabilities closely matched the experimental PHP values.

**Figure 15.** PHP pseudo-first-order kinetics biosorption.**Figure 16.** PHP pseudo-second-order kinetics biosorption.

Yet, the pseudo-second-order model offers a far more accurate description of the adsorption kinetics than the pseudo-first-order model, as evidenced by the consistently high R^2 values and the close agreement between calculated and observed q_e values. Although the pseudo-first-order model produced relatively high R^2 values, a good kinetic model should satisfy two criteria first one is a high coefficient of determination (R^2), while second one is a close agreement between the calculated and experimental equilibrium adsorption capacities (q_e). The pseudo-second-order model fulfills both criteria, whereas the pseudo-first-order model fails to accurately predict the equilibrium adsorption capacity for several

metals, particularly Zn and Cd [70]. The pseudo-second-order kinetic model is the best model for explaining the adsorption of Cd(II), Cu(II), Pb(II), and Zn(II) onto peanut-hull adsorbents because of the higher correlation coefficients and the improved agreement between calculated and experimental equilibrium adsorption capacities [68]. The pseudo-second-order model's supremacy indicates that the availability of adsorption sites and the interaction between the adsorbate and the adsorbent surface are the main factors influencing the adsorption rate. This behavior involves electron sharing or exchange between the metal ions and the functional groups on the adsorbent surface [73].

Table 6. Coefficients of sorption kinetics for Pb, Zn, Cu, and Cd removal using PHP and their correlation coefficient (R^2).

HMs	First Order				Second Order			
	$k_1 \text{ min}^{-1}$	$q_e \text{ cal mg g}^{-1}$	q_e	R^2	$k_2 \text{ min}^{-1}$	$q_e \text{ cal mg g}^{-1}$	q_e	R^2
Cd	0.12	95.30	90.43	0.99	0.005	96.29	90.43	0.99
Cu	0.10	89.86	92.89	0.99	0.005	98.84	92.89	0.99
Pb	0.11	92.85	93.14	0.97	0.006	98.58	93.14	0.99
Zn	0.10	90.77	89.93	0.99	0.005	95.45	89.93	0.99

3.8. Thermodynamic Analysis

Table 7 displays the thermodynamic parameters derived from the Van't Hoff analysis. For every metal ion under investigation, the computed values of ΔH were negative, ranging from -14.80 to $-22.49 \text{ kJ.mol}^{-1}$. This suggests that the adsorption of Cd(II), Cu(II), Pb(II), and Zn(II) onto the peanut hull-based adsorbent is an exothermic process [77].

Table 7. Thermodynamic parameters.

Metal Ion	ΔH° (kJ.mol^{-1})	ΔS° (kJ.mol^{-1})	ΔG° 283 (kJ.mol^{-1})	ΔG° 293 K (kJ.mol^{-1})	ΔG° 303 K (kJ.mol^{-1})	ΔG° 313 K (kJ.mol^{-1})	ΔG° 323 K (kJ.mol^{-1})	R^2
Cd(II)	-14.80	-67.19	4.21	4.88	5.55	6.22	6.90	0.825
Cu(II)	-21.18	-92.38	4.98	5.91	6.83	7.75	8.68	0.819
Pb(II)	-22.49	-95.97	4.67	5.63	6.59	7.55	8.51	0.842
Zn(II)	-17.72	-77.71	4.25	5.03	5.81	6.58	7.36	0.812

The experimental results, which demonstrate a general decline in adsorption effectiveness as the temperature increased from 10 to 40 °C, are consistent with the negative enthalpy values, which indicate that rising temperatures are generally detrimental to the adsorption process [78]. The order of ΔH magnitude was as follows: $\text{Pb}^{2+} > \text{Cu}^{2+} > \text{Zn}^{2+} > \text{Cd}^{2+}$. This suggests that compared to Zn(II) and Cd(II), the adsorption of Pb(II) and Cu(II) is linked to a larger enthalpic shift. Although the significance of particular interactions between metal ions and functional groups on the PHP surface cannot be ruled out, the very moderate amount of ΔH indicates that the adsorption process may entail primarily physical interactions [79]. A decrease in randomness at the solid-solution interface during adsorption is indicated by the negative values of ΔS , which varied from -67.19 to $-95.97 \text{ J mol}^{-1} \text{ K}^{-1}$. This could be explained by the immobilization of metal ions from the aqueous phase onto the adsorbent surface's active sites. The adsorbed metal ions may become more ordered after interacting with the PHP adsorbent's active sites, according to the negative entropy values [80]. The computed ΔG values were positive and rose as the temperature rose. This suggests that the adsorption process becomes less thermodynamically favorable at higher temperatures based on thermodynamic calculations based on the (K) values em-

ployed. The exothermic nature of the adsorption process is supported by the rise in ΔG with temperature, which is compatible with the negative ΔH readings [81]. Cd(II) had the lowest ΔH magnitude among the metals under investigation, while Pb(II) had the greatest. When compared to Cu(II) and Pb(II), the comparably lower ΔG values for Cd(II) and Zn(II) indicate relatively more favorable adsorption under the studied conditions. Nevertheless, the physicochemical characteristics of the metal ions and the experimental adsorption percentages should be taken into account in addition to the thermodynamic interpretation [82]. However, the thermodynamic study shows that there is a decrease in interfacial randomness and that the adsorption of heavy metal ions onto the PHP adsorbent is primarily exothermic. As the temperature rises, the adsorption becomes less favorable, indicating that lower temperatures are better for the adsorption process.

3.9. Investigation of Municipal Drainage Samples

Large and diverse amounts of pollutants from homes, hospitals, food, textile, paint, tanning factories, various mechanical and electronic industries, heavy industries, and other municipal and industrial sources greatly increase the pollution load in municipal drainage water [3]. Thus, the current study's findings showed that the effluent water of drainages had quantities of heavy metals (lead, zinc, copper, and cadmium) that were higher than allowed, with values of 2.78 ± 0.8 mg/L for Pb and 0.03 ± 0.0 mg/L for Cd.

The laboratory treatment unit was used to clean the studied drainage water samples of their heavy metal content. Untreated wastewater was added to the first tube at a volume of approximately 3 L, and then the system containing PHP immobilized with calcium alginate beads was operated at a rate of 2 L/h. After that, the total heavy metal content was examined from the third tube for collecting water samples after treatment (Figure 17).



Figure 17. Laboratory treatment unit.

When analyzing the water released from such drainages, the results show that there are minor variations in these concentrations' values. Nevertheless, all of these concentrations vanished after the highly turbid, contaminated water sample was treated with PHP beads through the treatment unit, which was very successful in removing the pollutants under investigation and turning it into clear water with virtually little turbidity.

The quantities of heavy metals in the understudied Ramadi municipal drainages before and after treatment with PHP beads are displayed in Table 8, along with the World Health Organization's acceptable limits for heavy metals in the effluent waters [83].

Table 8. HM concentration in understudy drainages before and after treatment by PHP.

Sites	HM Concentration in Drainage Raw Water (mg/L)				HM Concentration After PHP Treatment				WHO Permissible Limits (mg/L)			
	Pb	Zn	Cu	Cd	Pb	Zn	Cu	Cd	Pb	Zn	Cu	Cd
S1	2.78 ± 0.8	1.45 ± 0.6	0.66 ± 0.1	0.03 ± 0.0		Not detected						
S2	1.94 ± 0.2	0.98 ± 0.0	0.79 ± 0.1	0.02 ± 0.0		Not detected			0.01	5	2	0.003
S3	2.14 ± 0.3	1.22 ± 0.2	1.32 ± 0.7	0.02 ± 0.0		Not detected						

The Euphrates River's heavy metal contents are significantly increased by the water released by understudied drainages or urban water drainage systems. This demonstrates that before being released into the river, this method must go through a tertiary treatment procedure that lowers or mitigates the amounts of heavy metals to allowable limits. Therefore, maintaining a sustainable environment that respects the right of future generations to a decent life.

4. Conclusions

This study was focused on peanut hull powder biomass as a low-cost adsorbent for the removal of Pb, Zn, Cu, and Cd ions from aqueous solution. The sorption is affected by various parameters such as contact time, solution pH, size and dose of hulls, and temperature. The Langmuir adsorption isotherm model was better used to represent all of the experimental data, which are fitted using the Langmuir isotherm, since the RL is favorable and the correlation coefficient is more than 0.95. Consequently, the adsorption isotherms for every metal ion were fitted to the Freundlich n with a correlation coefficient of more than 0.88 and n values greater than 1. Based on the kinetic simulation, SEM analysis reveals the changes in the hull's structure before and after treatment with heavy metal ions via biosorption, encouraging the idea that the cross-linking of functional groups and metal ions may be responsible for the biomass's morphological alterations. Also, FTIR analysis supports the idea that the main mechanisms involved in the removal of heavy metal ions under study are both surface ion exchange and surface or intra-particle sharing between metal ions and functional groups, such as O–H, C=O and C–H stretching, hence indicating that adsorption of Zn^{2+} , Cd^{2+} , Pb^{2+} , and Cu^{2+} ions occurs largely through oxygen- and nitrogen-containing functional groups on the surface of peanut powder. Nevertheless, the designed laboratory unit was effective in treating unacceptable levels of heavy metals in some municipal wastewater of Ramadi City local drainages, providing a preliminary concept for designing a field treatment system that could be implemented in related large-scale projects, with the possibility of studying the regeneration of PHP beads more than once. The overall results indicated that *Arachis hypogaea* hull biomass is an easily, locally available, low-cost adsorbent and has a considerably high biosorption capacity. It may be treated as an alternative biosorbent for treatment of wastewater containing such heavy metal ions.

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