Kurdistan Journal of Applied Research (KJAR)
Print-ISSN: 2411-7684 | Electronic-ISSN: 2411-7706

Original Article

Enhancement of Phosphorus Sorption onto Peanut Shell Using Aluminum and Iron Oxide Coatings

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Submitted: October 9, 2023
Revised: November 6, 2023
Accepted: December 14, 2023

10.24017/science.2023.2.4

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Keywords: Peanut shell, Al-coated, Fe-coated, phosphorus, adsorption, isotherms equations.

Abstract: Phosphorus serves as a crucial nutrient for the organisms. Still, the discharge of high levels of phosphate concentrations into limited aquatic environments leads to the phenomenon known as eutrophication, which subsequently leads to the degradation of the overall water quality. In this study the capacity of the peanut shell, Al-coated and Fe-coated is determined by adding 10, 20, and 30, 40 mg P L-1 as (KH2PO4) to 1.0 g of each sample and shaking for 2, 6, 12, and 24 h and at pH ranged between 4.6, 5.2, 6.3 and 7.8. At the end of each period, the suspension was filtered and analyzed which presents the concentration of equilibrium P. The maximum P adsorption of 28.9±0.07 and 50.6±0.49% was recorded for 24 h of incubation and at pH (4.6) with an Al-coated peanut shell respectively. The findings of this study demonstrated that the adsorption of phosphorus (P) increased as the duration of incubation increased. At the same time, it decreased with a decrease in the pH of the solution. Furthermore, the outcomes indicated that the Freundlich model provided the best fit for the data, based on higher R2 values ranging from 0.9867 to 0.9938, in comparison to the range of 0.9659 to 0.9904 observed for the Longmuir model. These results suggested that peanut shells coated with Al can be used to remove high amounts of P in the solution. Also, the removal resulted from the physical adsorption of P rather than a chemical reaction.

1. Introduction

In recent years, modern technologies have been introduced to remove specific elements from various types of waste [1- 5]. The existence of an excessive quantity of diverse anionic compounds, such as phosphate, can give rise to challenges concerning water resources. [6].

In water bodies, phosphate exists in various forms, including orthophosphate, polyphosphate, and organic phosphate [7]. Phosphate plays a pivotal role as an essential element required for the proliferation of living organisms within the majority of ecosystems. However, releasing heightened phosphate concentrations into limited aquatic environments leads to eutrophication, subsequently causing a decline in water quality. This degradation primarily occurs due to oxygen depletion caused by the decay of algae. [8, 9].

Excessive concentrations of diverse anions, such as phosphate, can give rise to complications in water bodies. [6]. Orthophosphate ‹polyphosphate and organic phosphate are the popular types of phosphate in water resources [7]. Henceforth, the concentration of phosphate in discharges or watercourses that are released into the natural surroundings is overseen by governing bodies adhering to national and international standards for wastewater. These regulations stipulate that the permissible upper limits for phosphate range from 0.1 to 2 mg L-1 [10]. For instance, the United States, India, and Iran have prescribed discharge thresholds for phosphate, namely 0.51, 5, and 6 mg L-1 as P, respectively. Phosphate elimination has garnered significant attention since the latter part of the 1960s [6, 11].

Numerous techniques have been employed to extract phosphate from wastewater, which can be categorized into physical, chemical, and biological approaches [12]. Physical methods tend to be cost-prohibitive, such as those involving electrodialysis or reverse osmosis [9]. In the context of a biological treatment facility, the transfer of phosphate from the liquid to the sludge phase is imperative. However, the efficiency of this removal process typically does not surpass 30%, necessitating alternative technologies to eliminate the residual phosphate [13].

Nowadays, the appeal of chemical processes has diminished due to drawbacks such as additional expenses for the purchase and installation of dosing equipment, challenges associated with sludge management and disposal, maintenance costs, and the use of chemicals [11, 14]. Another approach that has garnered significant interest in the field of wastewater treatment is the utilization of biosorbents. They have proven to be effective in the elimination of various types of contaminants, including important anions such as phosphate [15], heavy metals [16], and polycyclic aromatic hydrocarbons [17]. In this process, the biosorbent is in contact with pollutants in aqueous solutions at specific conditions. Thus, the objective of this investigation was to conduct a comprehensive comparison between aluminum and iron-coated peanut shells in terms of their ability to remove phosphate through the process of adsorption. In order to achieve this objective, the impacts of key variables including pH level, initial phosphate concentration, and incubation time were assessed for both types of coated particles. Furthermore, various adsorption isotherm models were employed to ascertain the specific characteristics of the adsorption process.

2. Materials and Methods

2.1. Preparation of sorbent materials

The manual crushing and subsequent sieving of the peanut shell material facilitated the selection of the particle-size fraction measuring 1-2 mm for experimentation purposes. In order to eliminate any impurities, a quantity of 130 g of the sieved peanut shell was immersed in 500 mL of a 0.02 M solution of HNO_3 for 24 hours. The peanut shell was then subjected to five thorough rinses with deionized water before being subjected to oven drying at a temperature of 105 °C for 24 hours. This resulting material was subsequently divided into three distinct portions. The initial portion employed for the application of an Aluminum oxide coating (known as Al-peanut shell) was produced using the procedure outlined in the study conducted by Edwards and Benjamin, 2009 [18].

In short, a solution of 0.5 M Aluminum (III) nitrate nonahydrate (Al (NO3)3.9H2O) was prepared by dissolving it in deionized water with a volume of 200 ml. The pH of the solution was subsequently adjusted to 11.1 by gradually adding concentrated Sodium hydroxide (NaOH) to the solution while simultaneously stirring it. Following this, the resulting solution was poured onto a collection of 60 grams of peanut shells that were placed on a stainless steel plate. The ratio employed was 60 grams of peanut shell per 200 mL of Aluminum (III) nitrate solution. The mixture was then subjected to heat by placing it in an oven at a temperature of 105 degrees Celsius for a duration o for the second portion, another 60 grams of peanut shell was utilized for the application of an Iron oxide coating (known as Fe-peanut shell) and was prepared using the method outlined in the research conducted by Han, et al. 2009 [19]. Briefly, a solution of 0.5 M Fe (NO3)3.9H2O was prepared in deionized water with a volume of 200 mL. The pH of the solution was subsequently adjusted to 11.13 by the addition of NaOH, as described previously.

Following this, the mixture was subjected to heating in an oven at a temperature of 60 0C for 48 hours. Subsequently, the two coated materials were subjected to multiple washes utilizing deionized water to eradicate any unfastened hydroxide, until the flowing water turned transparent.

Ultimately, the cleansed materials were subjected to a desiccation operation at a temperature of 105 0C for a period of 24 hours. These dried materials were then stored in glass bottles and were thus prepared for usage [27]. The third portion of the peanut shell remained uncoated and served as a reference material.

2.2. Phosphorus release experiment

Dried ground samples weighing 1.0± 0.2 g were introduced into a succession of acid-washed plastic centrifuge tubes with a capacity of 100 mL, along with 50 mL of ultrapure water. The aforementioned centrifuge tubes were sealed and positioned within an end-end shaker operating at a velocity of 100 revolutions per minute for 2, 4, 8, 16, and 24 hours. Then, the samples were filtered by Whatman 42 µm filter paper. The filtrates were kept in a refrigerator until the day of analysis. A spectrophotometer (SHIMADZU UV-1700 PharmaSpec UV-VISIBLE) was used to analyze the P concentration.

2.3. Phosphorus Sorption Index (PSI) experiment

A batch equilibrium technique was used to determine phosphorus sorption. Dried specimens (2.0± 0.2 g) were introduced into acid-washed centrifuge containers comprising 50 mL of the phosphate solution (KH2PO4) that encompassed 70 mg p L-1 [20]. The centrifuge bottles were capped and placed in a shaker at 100 rpm for selected time intervals including (2, 6, 12, and 24h) at room temperature (20±2 0C), each in triplicate, as well as a control sample [20]. The samples were filtered by Whatman 42µm filter paper, put into the 50 mL screw cap centrifuge tube, and kept in a refrigerator until analyzing day. The filtrates were analyzed by a spectrophotometer for the P concentration. The Phosphorus Sorption Index (PSI) was calculated as described by [20]:

$$PSI(l.kg) = \frac{x}{log\ c} \qquad (1)$$

Where:
X= P sorbet (mg P/kg) after shaking time
C= P concentration (mg l^{-1}) at equilibrium after shaking time.

2.4. Effect of pH on P adsorption

Adsorption experiments were carried out at different pH levels and with a consistent concentration of P (70 mg l^{-1}). The samples' pH was regulated in the range of 4.6 to 7.6 by the addition of a small amount of 0.1 M NaOH or 0.1 M HNO3. Following 24 hours of agitation, the samples were subjected to centrifugation and filtration [28]. The pH glass electrode was employed to ascertain the equilibrium pH of the suspension. P adsorption was assessed using a spectrophotometer, employing the same methodology as previously described.

2.5. Isotherm experiment

A study was carried out to investigate the absorption of phosphorus (P) in peanut shells under laboratory conditions. The experiment involved three replicates from each of the three peanut shells, and a time-dependent adsorption isotherm method was employed, as detailed in reference [21]. Dried specimens (1 g, 2 mm) were introduced into distinct 50 mL containers for centrifugation that had been cleansed with acid. These containers were accompanied by 25 ml of the phosphate solution (KH2PO4), which had varying concentrations of 10, 20, 40, and 80 mg/L, with each concentration being replicated. The centrifuge bottles were capped and placed in an orbital shaker at 100 rpm for 24 h at room temperature (22- 25°C). The suspension was left for an hour, filtered by Whatman 42μm filter paper, put into the 50 mL screw cap centrifuge tube, and kept in a refrigerator until the analyzing day. The filtrates were analyzed for dissolved P using the same instrument as was used in the PSI study. Total adsorbed P (ΔPt) was calculated by the following equation:

$$\Delta P = Pint - \ Pf \qquad (2)$$

Where:
Pint = P concentration at equilibrium (mg/L)
And Pf = final P concentration after an hour of equilibration (in mg/L).

The models of adsorption isotherms can be employed to determine the distribution of P between the solid and liquid phases using a linear function [22]. Consequently, in the present investigation, the Freundlich and Langmuir adsorption isotherm models, which are widely used in linear format, were utilized to examine the adsorption of P in the analyzed specimens. The mathematical expressions for the two aforementioned models are provided in equations [3] and [4]. The models for adsorption isotherms can be employed to establish the partitioning of P between the solid and liquid phases, utilizing a linear function as suggested by Tariq, et, al [21]. In this particular investigation, the Freundlich and Langmuir adsorption isotherm models, which are widely employed, were utilized in their linear formats to explore the adsorption of P in the samples under examination. The two models' general forms are depicted in equations.

2.6 Freundlich isotherm model

$$log\frac{x}{m} = logK + \ \frac{1}{n}\ logc \qquad (3)$$

Where:
x = mass of adsorbate
m = mass of adsorbent
c = equilibrium concentration of adsorbate in case of experiments made with an aqueous solution in contact with a dispersed solid phase (dissolved species/adsorbed species). K and n are constants for a given adsorbate and adsorbent at a given temperature.

2.7. Langmuir isotherm model

$$\frac{1}{q_{eq}} = \frac{1}{q_{max}\,k_l\,c_e} + \frac{1}{q_{max}} \qquad (4)$$

Where:

q_{eq} is the adsorbent at equilibrium (mg /g), $q_{max}$is the maximum adsorption capacity (mg g^{-1}). KL is an equilibrium constant (l mg^{-1}) related to energy of adsorption which quantitatively reflects the affinity between the adsorbent and adsorbate and Ce is the equilibrium concentration of the adsorbate in (mg l^{-1}).

3. Results and Discussion

3.1. Effect of pH values on phosphorus adsorption

The adsorption experiment was performed at different pH levels while maintaining a consistent P concentration. The samples' pH was controlled within the spectrum of 4.6 to 7.6. The extent of adsorption is presented in table 1. Ionic exchange is influenced by the pH of a solution. It is similar to Deutsch et al., [23] who reported that pH influences the P adsorption process. In every instance, the percentage of P that is adsorbed exhibits a decline when the pH value is increased. The elevation in the pH value from 4.6 to 7.6 demonstrates a decline in the advancement of cations residing on the exterior of the particles. This reduction in the affirmative charge present on the surface of the particles leads to a diminished electrostatic interaction between the adsorbent and the adsorbate. This observation aligns with the findings of Tariq, et al. 2021 [21], who examined the rate at which phosphorus is adsorbed in calcareous soils. In addition, the OH- ions in the solution are increased. Resulting in the competition between OH- and PO4- was created. Therefore, a small amount of phosphorus was adsorbed. The observed decreases in the electrostatic interaction resulted in a substantial reduction in the proportion of adsorbed phosphorus (P) for the Peanut shell, Al-coated, and Fe-coated particles. Consequently, the alteration in pH values from 4.6 to 7.6 corresponds to a decrease of approximately 19.9 to 6.18, 50.6 to 35.3, and 40.2 to 22.9 of the proportion of adsorbed P for the Peanut shell, Al-coated, and Fe-coated particles, respectively.

Table 1: Effect of pH values on the phosphorus adsorption at Peanut shell, Al-coated, and Fe-coated particles.

Type	**Controlled pH**	**Adsorbed P %**	**±SD**
Peanut shell	4.6	19.9	0.07
	5.2	18.5	0.09
	6.3	14.7	0.30
	7.6	6.18	0.66
Al coated	4.6	50.6	0.49
	5.2	47.1	0.08
	6.3	41.5	0.26
	7.6	35.3	0.55
Fe coated	4.6	40.2	0.17
	5.2	35.7	0.11
	6.3	29.5	0.73
	7.6	22.9	0.20

3.2. Phosphorus releasing

Phosphorus release experiments were carried out at zero P concentration. The pH value was 6.4 at room temperature. In all cases, P was not released into the solution. Even though the contact time during shaking was increased from 2 to 24 hours, the sample did not release any phosphorus.

3.3. Phosphorus sorption index (PSI) experiment

Phosphorus sorption index experiments were carried out at initial P concentrations equal to 70 mg l^{-1} with different shaking times (2, 6, 16, and 24). The initial pH value was 6.1. The average P adsorbed in solution ranged from 42.1 to 67.3 mg l^{-1} as shown in table 2. This means peanut shells could be used as bio-sorbents to remove P in the solution. P adsorption was lower for Peanut shells than Al-coated, and Fe-coated particles in all cases. There is a significant difference between uncoated and coated peanut shells. The high PSI value means the greater potential of the peanut shell to adsorb phosphorus from the solution. The ratio of P adsorption between Al-coated and peanut shells were 17.7: 4.2 and 14.4: 5.4 for the shortest and longest shaking time respectively. On the other hand, this ratio between Fe-coated and peanut shell was 3.1:1 and 2.1:1.1 for the same shaking times as shown in table 2. In other words, the Al-coated ability of the adsorption of phosphorus was higher than Fe-coated. It supports the fact that P interaction with Fe is weaker than Al particles. In addition to this result, the amount of P adsorbed by Al-coated is greater than that of Peanut shell particles. The amount of P remaining in the solution was decreased with increasing incubation time. This is similar to Jarvie et al [18]. Increasing incubation time allows the peanut shell to adsorb a higher amount of phosphorus.

Table 2: Phosphorus adsorption on peanut shell, Al-coated, and Fe-coated at different incubation times.

Type	Incubation time (h)	P adsorbed mg l-1	±SD
Peanut shell	2	4.20	0.51
	6	7.01	0.54
	12	8.76	0.61
	24	10.8	0.28
Al-coated	2	17.7	0.38
	6	21.3	0.64
	12	25.7	0.61
	24	28.9	0.07
Fe-coated	2	12.4	0.15
	6	15.5	0.39
	12	17.8	0.32
	24	20.9	0.21

Table 3: The parameters of Freundlich and Langmuir models onto peanut shells, Al-coated particles, and Fe-coated particles.

Type	Freundlich's equation		Langmuir's equation	
	KF	1/n	RL	KL
Peanut shell	4.10	1.26	0.62 0.45 0.29 0.17	0.06
Al-coated	1.45	1.21	0.27 0.16 0.08 0.04	0.27
Fe-coated	2.01	1.38	0.76 0.62 0.45 0.29	0.03

3.4. Adsorption isotherms

Adsorption of P on Peanut shells rather than Al-coated and Fe-coated particles was investigated. The persistent adsorption of P over time is expected to result in a significant alteration in the adsorption isotherm [21]. Both Freundlich's equation (1) (Figure 1) and Langmuir's equation (2) (Figure 2) were successfully fitted to the experimental data with a coefficient of determination (R2) greater than 0.96. The Freundlich and Langmuir parameters for the adsorption of P on peanut shells, Al-coated, and Fe-coated particles are presented in table 3. In all instances, the Freundlich equation provided better fits compared to the Langmuir equation, as evidenced by the higher values of R2 ranging from 0.992 to 0.987 to (0.966- 0.990) for the Langmuir. These outcomes were derived from the equations they developed based on Figures 1 and 2.

Figures 1 and 2 demonstrate that the data, when linearized, adhered to the Freundlich and Langmuir model, resulting in the formation of linear relationships. The phenomenon of phosphate sorption demonstrated an upward trend as the concentration of phosphate in the liquid phase increased. It is noteworthy to mention that the sorption isotherms were not inclined to become less steep when confronted with higher concentrations of phosphate. In other words, phosphate sorption increased with increasing liquid-phase phosphate concentration; the isotherms did not become flatter at higher phosphate concentrations (Figures 1 and 2). This outcome aligns with the findings of Nazarian et al. [25], who concluded that the Freundlich and Langmuir model proved to be the most suitable fit for the adsorption of P in lanthanum oxide supported on activated carbon. As anticipated, the outcome aligns with initial predictions. The Freundlich constant for the Peanut shell is 2.8 times greater than that for Al-coated. Similarly, for Iron, the value is 2.04 times larger than that for Fe-coated as indicated in table 3. Nevertheless, the value of 1/n remains almost unchanged for Peanut shells and Al-coated particles. Specifically, Peanut shells demonstrate an average value of 1.26, whereas the corresponding value for Al-coated is 1.21. This value for Fe-coated is 1.38, which is greater than the Peanut shell and Al-coated particles.

In the context of the Langmuir model, the adsorbent's affinity towards the adsorbate is indicated by KL. Strong binding is suggested by high values of KL. The frequent occurrence of KL signifies the resemblance between adsorbent colloids and P ions, which serves as a measure of the adsorbent's adsorption capacity [26]. In the present investigation, the KL values varied from 0.03 mg l^{-1} for Fe-coated, 0.06 for peanut shell, to 0.27 for Al-coated particles as shown in table 3 and figure 2, which implies the advantageous circumstances for the adsorption of P through the utilization of peanut shell. A positive K-value indicates the spontaneity of the adsorption reaction at ambient temperature, while KL's significance indicates the extent of this spontaneous adsorption reaction [21]. The increased K-value corresponds to a higher degree of spontaneity and greater stability of the products, resulting in an increased retention of P [26]. The KL-value in table 3 demonstrated a marked increase in the case of the Al-coated particles in comparison to the Fe-coated particles and peanut shell particles as indicated in table 3. Furthermore, a noteworthy disparity was observed among all the peanut shell particles under study, indicating that the adsorption of peanut shell P experienced enhancement as the applied amount of P and contact time were augmented. The RL means the ability of particles to adsorb P in the solution. The range of RL values for peanut shells was observed to be between 0.17 and 0.62, while for Al-coated it ranged from 0.04 to 0.27, and for Fe-coated and peanut shell particles were 0.29 to 0.76 at 24 hours of contact time. Additionally, the RL values indicate that the P adsorption showed higher variability at lower concentrations compared to higher concentrations, regardless of the contact time. This variability can be explained by a decrease in the particle's ability to adsorb P as the phosphate concentration increases [21]. This occurrence is associated with the saturation of highly attractive attachment sites on peanut shell particles and the subsequent increase in electrostatic barriers due to the fixation of negatively charged phosphates onto the particle surface [22]. However, this result means that Al-coated has a higher capacity to retain P compared to Fe-coated and peanut shell particles. This outcome corresponds with the study conducted by Behbahani et al., [22], which concluded that the aluminum electrode exhibits a greater capability for phosphorus removal when compared to the iron electrode under identical operational conditions.

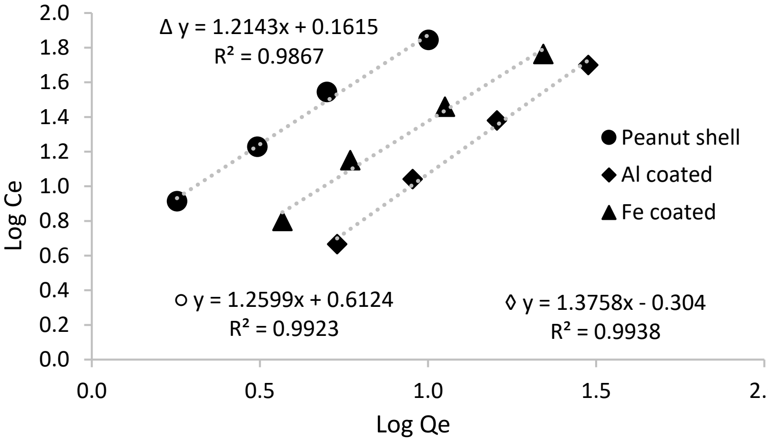

Figure 1: Adsorption of P on (○) Peanut shells, (◊) Al-coated, and (◊) Fe-coated particles according to the Freundlich. equation.

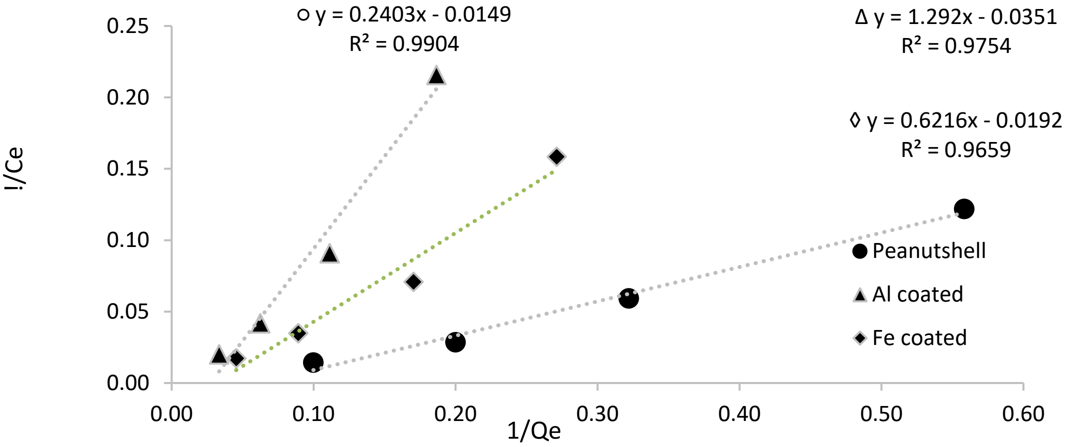

Figure 2: Adsorption of P on (○) Peanut shells, (◊) Al-coated, and (◊) Fe-coated particles according to the Langmuir equation.

4. Conclusions

Time-dependent and range of different pH for P removing in solution were investigated in this study using peanut shells, Al-coated and Fe-coated. The findings demonstrated that the adsorption of P from the solution augmented as the incubation time lengthened and the pH of the solution declined. Optimal adsorption was observed when the incubation time was at its maximum, the surface was coated with aluminum, and the pH reached its lowest value. This indicates that peanut shells, Al-coated, and Fe-coated had a high capacity for removing P in the solution. Moreover, Al-coated and pH 4.6 had the highest adsorption capacity than peanut shells and Fe-coated and pH 5.2, 6.8, and 7.3 respectively. Furthermore, the Freundlich isotherm model was found to be more appropriate for characterizing the P adsorption behavior in the solution across all investigations. It is also better fitted to peanut shells than Al and Fe- coated materials. Increasing in P adsorption corresponded with increasing concentration of P, contact time, and decreasing pH value. Further studies are recommended to investigate the optimum ability of coated peanut shells to remove phosphorus. These results indicate that the aluminum and iron-coated peanut shell could be utilized as an economical sorbent for wastewater cleanup from phosphorus.

Data availability: Data will be made available on request.

Conflicts of interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Funding: The authors did not receive support from any organization for the submitted work.

Reference

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