

Study of Biomass Bottom Ash Efficiency as Phosphate Sorbent Material

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Abstract

Excessive richness of nutrients in water bodies such as rivers, lakes and ponds lead into deterioration of aquatic life as a results of dense growth of algae. Phosphate is one of the main nutrients that should be controlled to prevent this serious issue. Utilizing low cost material as a phosphate sorbent is offering a treatment method characterized as a sustainable solution. In this study the efficiency of biomass bottom ash BBA as phosphate sorbent material from aqueous solution is investigated. Batch experiments were undertaken, in which a particular mass of BBA was brought into contact with the phosphate solution. The experiments studied the influence of pH (different phosphate solutions were prepared with pH range 4 to 8), temperature (adsorption capacity measured at the temperature range of 10 to 30 °C), and contact time. In addition, the adsorption isotherm models were also applied to better understand the mechanism of phosphate sorption by BBA. The results revealed that the bonding between the cations (BBA surface) and anions (phosphate solution) is significantly affected by the pH of the solution. BBA presents an excellent phosphate sorption, especially, at low pH value and temperature around 20 °C. The method of this research can be adopted as a followed strategy for examination the capability of selected material for phosphorus removal from wastewater.

Keywords: Adsorption Isotherm Models; Biomass Bottom Ash; Filter Media; pH; Phosphate Sorption; Wastewater Treatment

1. Introduction

Phosphorus is one of the essential nutrients that contribute in eutrophication the water bodies such rivers and lakes [1, 2]. Eutrophication or also called algal blooms is the process of extraordinary growth of the algal as a results of nutrients release into the water bodies. The Eutrophication play a negative role by preventing the sunlight from reaching the aquatic vegetation; and decrease the dissolved oxygen as a result of decomposition of their organic matter that lead to present of adverse effects on the aquatic life. The diversity of the higher forms of the aquatic life will be reduced because of the poor conditions of the water system that caused by eutrophication such as anoxic, acidic, detrimental conditions [3]. Recently, many regulations were enacted to protect the water bodies from the nutrients; specifically, the phosphorus. Water Framework Directive (WFD) which is legislated by European Union EU is one of these regulations that tightening the nutrient discharge into water bodies [4]. All over the world, a significant attention was paid to prevent the effluent of nutrients into the ecosystems to avoid their adverse effects. Therefore, seeking for new techniques to control the nutrients concentrations became significantly required.

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Phosphorus is released into the aquatic environment as a result of many human activities [5]. Sewage effluent represent one of the main P sources that release into the water bodies; Bowes et al [6] stated that the most significant source of P in UK is come from sewage effluent and he mentioned that up to 70% of P contributed by sewage discharges. Installation of P removal technology in large wastewater treatment plants become quite common [7]. However, receiving P discharges from small wastewater treatment plants is often treated less rigorously; and the potential adverse effects of phosphate that results from small communities may be underestimated [8].

The recent advances in the process of phosphate removal [9, 10] suggested to combine a techniques such as filtration and tertiary-ballasted flocculation with metal salt dosing to control the phosphate concentration at a permissible limit. However, these suggestions still have high cost and sophisticated to be implemented in wastewater facilities for small communities. The most important aspect in the process of wastewater treatment is to eliminate the contaminant from it and release it as safe effluent into the aquatic environment. However, achieving low cost wastewater treatment technologies became vital element in the plant design [11]. Recently, utilize of active filter media to remove the contaminants from wastewater has been considered for use in treatment units of the wastewater plants. [12-14]. Process of selection the active filter media is a significant step that should be considered in the stage of filtration system design. The proper selection of active filter media will help to achieve the required quality that proposed by water environmental bodies. Based on literature review, the phosphate capture chemically by phosphate sorbent materials PSM is the dominant mechanism [15-17]. The PSMs which they contain Fe/Al hydroxides or easily soluble Ca/Mg compounds are showed significant efficiency to retain the phosphorus [18]. Various sorts of PSMs were investigated for phosphate removal; but most these filter materials investigated at high phosphate concentrations in wastewaters [19-21].

Batch experiments were conducted to introduce the estimates for phosphate retention by BBA. In a typical batch test, a fixed amount of substance comes into contact with the phosphate solution [22]. Commonly, the sorption isotherm models are used in the studies of phosphate sorption by PSMs to evaluate the phosphate sorption capacity and the nature of affinity [15].

This paper seeks to demonstrate that BBA can act as a filter media for the removal of soluble phosphate in an aqueous solution. In addition, the paper also aims to assess the fluctuation of the solution's pH value on the sorption process according to the experimental work and the mathematical model. Phosphate control is one of the essential action that should be taken to maintain the aquatic life in water bodies. This work introduce a sustainable solution by control the waste by waste and suggest a method for the researchers to be followed in regard the PSMs selections.

2. Materials and Method

2.1. Materials

The BBA sample was collected from the Fiddlers Ferry Power Station in the North-West of England; the BBA is result of burning different types of timber. Sieve analysis has been performed to identify the BBA particle size in the collected samples. According to the results in table 1 there are different masses of BBA retained on sieve 10, 16, 30 and 50. It is noticeable the largest amount has been retained on sieve 10. Then sequentially the quantity of BBA sample has decreased at sieves 16, 30 and 50 at percentage 28, 12 and 8.5% respectively. The BBA sample offers a wide range of particle sizes. However, the dominator particle size was 1-2 mm.

Table1. The results of sieve analysis for BBA sample

Sieve No.	Diameter (mm)	Mass of sample (g)	Percent retained (%)	Cumulative retained (%)	Percent finer (%)
4	4.75	0.00	0.00	0.00	100.00
6	3.35	14.00	2.00	2.00	98.00
10	2.00	304.5	43.50	45.50	54.50
16	1.18	196.00	28.00	73.50	26.50
30	0.60	84.00	12.00	85.50	14.50
50	0.30	59.50	8.50	94.00	6.00
Pan		42.00	6.00	100.00	0.00
Sum = 700					

X-ray fluorescence spectrometer XRF was performed to characterize the chemical composition of the BBA as shown in table 2. The key target of this test is to determine if the materials are consisting of the oxides that tend to retain the phosphate ions such as Al₂O₃, Fe₂O₃, CaO and MgO. In addition, to inspect if the materials contained an oxide that indicates a hazardous level of heavy metals.

Table 2. X-ray fluorescence analysis of the composition of BBA

Composition	Weight %
Al ₂ O ₃	7.168
SiO ₂	12.49
K ₂ O	2.973
CaO	7.615
Fe ₂ O ₃	4.397
MgO	0.451
CHO	63.95
ZrO ₂	0.007
ZnO	0.005
finer	0.939

In this experimental work a phosphate aqueous solutions at concentration of 10 mg/l was prepared as a source of phosphorus. The aqueous solutions of phosphate was prepared by dissolving potassium dihydrogen orthophosphate (KH₂PO₄) in deionized water. According to several researchers the concentration of Phosphorus in municipal wastewaters is vary [23-25]. However, Jenkins et al. [26] stated that the concentration of total phosphorus (organic and inorganic) in municipal wastewater is 10 mg/l; bacterial action will cause to decomposed most of the phosphorus into orthophosphate. Based on the above literature review, the aqueous solution was prepared at a concentration of 10 mg/l to simulate the highest expected concentration. In this work the modification of pH for aqueous solutions has required. The pH was adjusted by adding either hydrochloric acid (HCl) or sodium hydroxide (NaOH).

HACH LANGE DR 2800 spectrophotometer was used to measure the phosphate concentration in this experiments based on Amino Acid method, which helps to measure the phosphate concentration at range 0.2 to 30 mg/l.

2.2. Method

Batch experiments were conducted to obtain an initial understanding of how the BBA can be used as a PSM. Firstly, a specific volume of the phosphate solution was brought into contact with FBA mass in one-liter capacity flask to determine the contact time of optimal removal efficiency. The experiment was performed at room temperature so as to maintain the environmentally relevant condition. The material soaked in the solution did not show any upward buoyancy at material/solution ratio of 1/1.74. The suspension was shaken at a constant speed (100 rpm) for 2 min to allow all the surface area to come in contact with solution ions. Then it was allowed to stagnate over the course of the experiment to prevent the effect of mixing on the sorption results. In the beginning, the samples were collected at minor intervals; then the collection time was increased with the progress of the experimental time. Therefore, the samples collection times were 5 min, 15 min, 30 min, 1 h, 2 h, 4 h, 8 h, 16 h, and 24 h. The Batch experiments conducted for BBA samples in two situations. Firstly, the BBA samples as raw material without any pre-treatment. Secondly, BBA samples were washed several times with deionized water until run-off was clear. Then, BBA samples were dried at 110 °C.

Secondly, phosphate solutions were brought into contact with different BBA masses at various temperature ranges (10 °C, 20 °C and 30 °C) to identify the variation in phosphate sorption behavior according to isothermal models. The temperature was controlled by placing the flasks in a water bath. In the previous batch experiment, BBA mass of 23 g and a 40 ml solution were used to achieve a material/solution ratio at 1/1.74. Hence, to obtain the required data for isothermal models, the amount of solution was kept at 40 ml, and the BBA masses were changed to 16 g, 23 g, 30 g, and 37 g.

In addition, the phosphate solutions with pH 4, 5, 6, 7 and 8 were prepared, and the absorption of BBA at each pH value of the solution was measured at the same contact time and temperature. The typical pH of municipal wastewater before treatment is 6.5 to 8 [26]. However, the pH range was 4 to 8 to present a broad perspective on the pH variation and its influence on phosphate sorption by BBA.

All samples were filtered through a 0.45 µm filter paper to exclude the suspension particles that may interfere in the scattering of light when applied to the spectrophotometer. All experiments were carried out in triplicate, and the average value was used. An additional flask containing 40 ml of phosphate solution was run as a blank over all experiments for quality assurance.

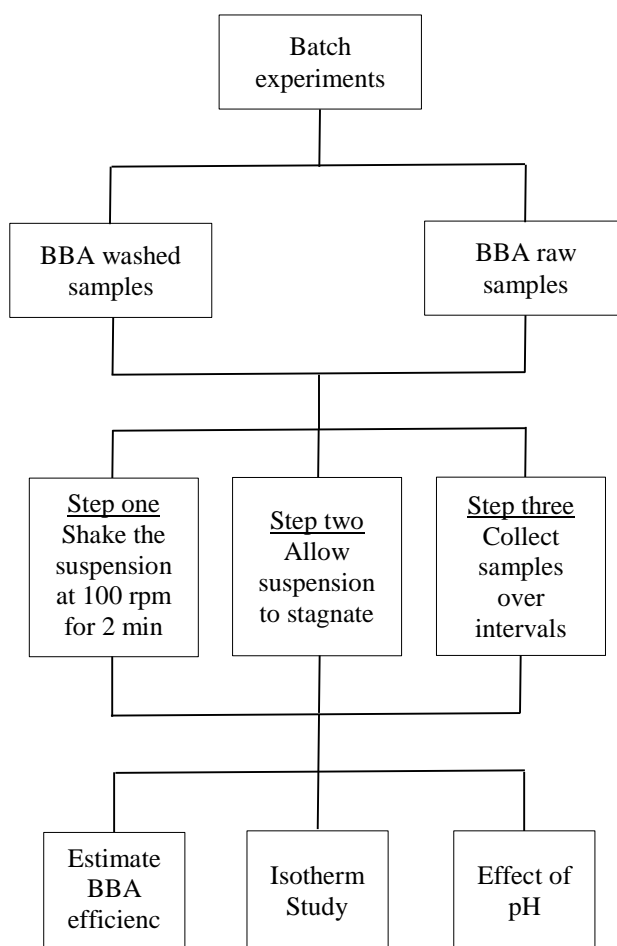


Figure 1. Schematic diagram for research methodology

2.3. Isothermal Adsorption Models

The equilibrium isotherm models are a significant tool for interpreting the adsorption mechanism in any system. Adsorption isotherm represents the magnitude of a solute adsorbed at a constant temperature and its concentration in the equilibrium solution. In addition, the applicability of the adsorption process as a complete operation was assessed through the physicochemical data provided by the adsorption isotherm model [27]. Several isotherm models are available for analyzing the data of experimental adsorption equilibrium. In this work, the BBA sorption characteristics were analyzed according to the Langmuir and the Freundlich models, which are widely used to describe phosphate retention isotherms for natural materials. The obtained data of the phosphate equilibrium concentration was fitted to the Langmuir and Freundlich isotherm models, as per the following isotherm equations:

The Langmuir equation is as follows:

$$q_e = \frac{Q K C_e}{1 + K C_e} \tag{1}$$

Where q_e is the mass of adsorbed per mass unit of adsorbent (mg/g). C_e is the equilibrium concentration of adsorbed (mg/L). Q is the maximum mass adsorbed at saturation conditions per mass unit of adsorbent (mg/g), and K is the empirical constant with a unit (l/mg) [28]. The Langmuir equation in linear form is given as follows:

$$\frac{1}{q_e} = \frac{1}{Q} + \frac{1}{K \cdot Q} \cdot \frac{1}{C_e} \tag{2}$$

The constants, K and Q , relate to the energy of adsorption and maximum adsorption capacity. The Freundlich equation is stated below:

$$q_e = K_f C_e^{1/n} \tag{3}$$

Where q_e is the mass of adsorbed per mass unit of adsorbent (mg/g), C_e is the equilibrium concentration of adsorbed (mg/L), and K_f and n are Freundlich constants, which correspond to the adsorption capacity and adsorption intensity, respectively. The equation can be linearized as below:

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

The application of isotherm equations becomes valid when the obtained data is fit to the model. In some cases, the model cannot analyze the findings. Langmuir isotherm model was suggested for homogeneous adsorption, and it is supposed to be a uniform adsorbent surface with energetically same sorption sites [29]. On the other hand, the Freundlich model describes equilibrium on heterogeneous surfaces.

3. Results and Discussion

3.1. Estimate BBA Efficiency

As mentioned in the method section the BBA samples has been brought in contact with phosphate aqueous solutions at concentration 10 mg/l. All Batch experiments were conducted at room temperature and the pH of aqueous solutions was maintained at 6.5. Figure 2 showed the capability of BBA for phosphate removal when it's raw material RBBA and after washing it WBBA.

RBBA and WBBA remove 79% and 90%, respectively from the total phosphate at the end of the experiment. It is noticeable that both samples have the same removal pattern; where the removal was steep at the beginning. Specifically, for first 2 hours of the experiments (55% and 59.5% for RBBA and WBBA). It is clear that the phosphate uptake was slow in rest of contact time. However, the removal period between 2 to 8 hours showed better removal efficiency in comparison with removal from 8 to 24 hours. Based on the outcomes, the process of phosphate uptake onto BBA consist of three phases (fast, moderate and slow phase). It is expected that the fast uptake it take place onto the external surface for the BBA samples, and the uptake became slower in lately phases because the phosphate will start to transfer from the external surface into the pores.

The removal efficiency for both samples in fast phase was close to each other. While, in the moderate phase the removal efficiency for WBBA was better than RBBA. Along the moderate phase period the phosphate removal was 21.3% and 15.8% for WBBA and RBBA, respectively. Finally, in the slow phase the removal efficiency for both samples back to be close. The run off of the BBA washing indicated appearance of some debris; it might be the debris removal allow the phosphate solution to reach more deep pores that increase the chance of contact. However, the removal efficiency for both samples decreased in the final phase as a results of transfer the phosphate ions from wide pores into microspores and the concentration of phosphate aqueous solution became very low.

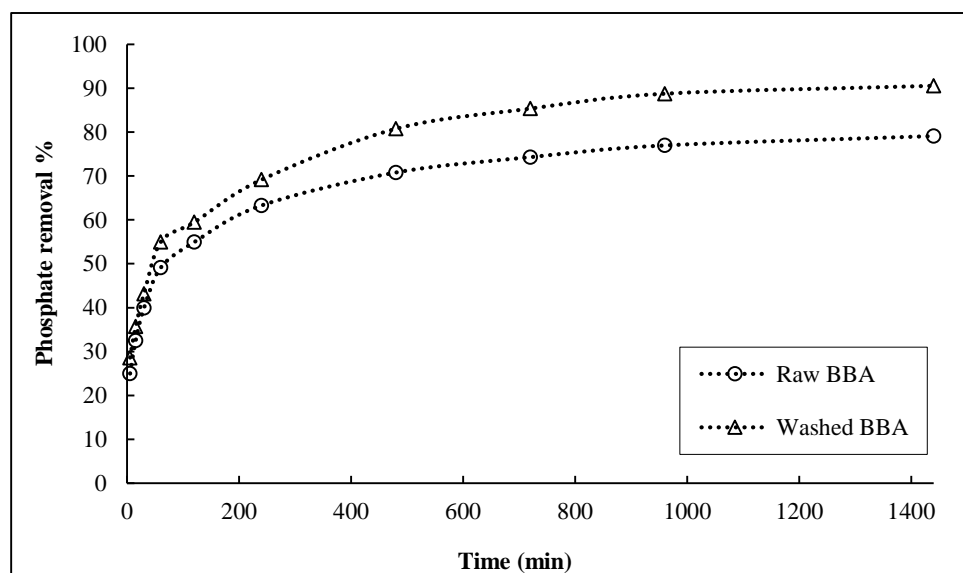


Figure 2. Influence of contact time on P sorption by RBBA and WBBA (Solution concentration = 10 mg P/l, pH = 6.5, contact time = 24 hr)

The contact time for the rest of the batch experiments was set at 8 hours because after this time the removal efficiency became very low.

3.2. Adsorption Isotherm Study

The behavior of phosphate adsorption at different temperatures is illustrated in Figure 3, which represents the function of equilibrium of phosphate concentration (C_e) vs. mass of adsorbed per mass unit of adsorbent (q_e). The results revealed a similar pattern of phosphate adsorption by BBA at different experimental temperatures. However, the adsorbed

phosphate at 20 °C looked much better than the adsorption at 10 °C and 30 °C. Moreover, the lowest value of adsorbed phosphate at 20 °C (0.025 mg P/l) seemed to be the highest than the adsorption at 10 °C and 30 °C.

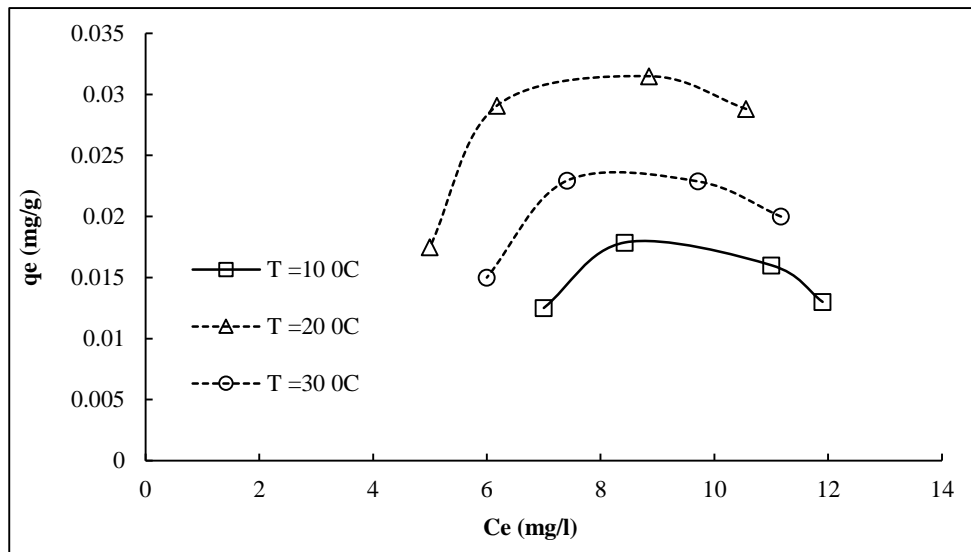


Figure 3. Adsorption isotherms of phosphate on furnace bottom ash

It is hard to deny the fluctuation of temperature in real life; the sharp decrease or increase in temperature will negatively affect the phosphate sorption via BBA. The removal capacity can be maintained by increasing the quantity of BBA. Since BBA is a waste material and freely available, it is required to increase the units' number of the treatment facility to accommodate the increase of BBA quantity. Consequently, this might have a negative impact on the footprints of the treatment facilities.

Langmuir and Freundlich isotherm models were applied to describe the distribution of ions between the liquid phase PO4-3 and the solid phase BBA. The linear form of the Langmuir equation represented in Equation 2 leads to the redrawing of the adsorption isothermal relationship as the inverse of equilibrium concentration 1/Ce vs. the inverse of the mass of adsorbed per mass unit of adsorbent 1/qe, as shown in Figure 4.

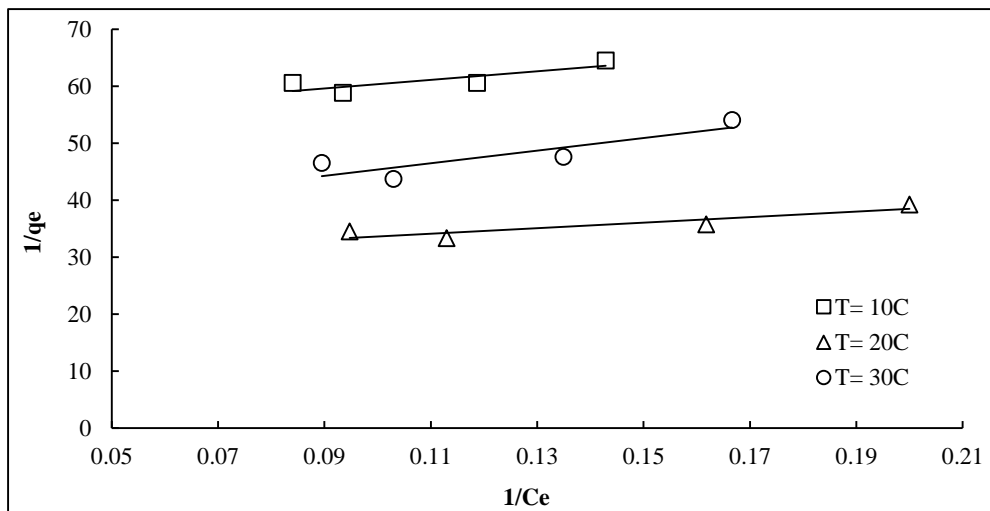


Figure 4. Langmuir isotherm model (1/Ce vs. 1/qe) at initial pH 6.5, contact time 8 hours

The determination of the maximum phosphate adsorption by the BBA surface indicates the BBA saturation point. The constant (Q) in the Langmuir equation represents the maximum adsorption by the surface. This constant is obtained from the interception of the plot in Figure 4. As shown in Table 3, the maximum adsorption capacity for BBA at 20 °C can be estimated to be 0.035 mg/g. This result indicates that the BBA had a higher affinity for P at 20 °C in comparison with the values at 10 °C and 30 °C, where the maximum adsorption capacity was 0.02 and 0.03 mg/g, respectively.

Table 3. Langmuir and Freundlich isotherm models' parameters for P adsorption on furnace bottom

Temperature (°C)	Langmuir			Freundlich			
	Q	K	R ²	R _L	K _f	n	R ²
10	0.020	0.7	0.69	0.11	0.013	7.77	0.64
20	0.035	0.6	0.83	0.12	0.020	5.58	0.75
30	0.030	0.3	0.77	0.22	0.011	3.75	0.69

The coefficient R² indicates that the phosphate adsorption by BBA gives a good fit with the Langmuir model as seen from the values of regression coefficients presented in Table 3. The data well-obeyed the Langmuir model at 20 °C, where the R² value was 0.83. The slope of the plot of 1/q vs. 1/C gives the value of constant K. The K and the initial phosphate concentration (C_i) were applied to estimate the affinity between the P and BBA using a constant called separation factor or equilibrium parameter (R_L) [27], which can be expressed as follows:

$$R_L = \frac{1}{1 + K C_i} \tag{5}$$

The values of R_L between 0 and 1 indicate favorable adsorption while R_L values over 1 indicate unfavorable adsorption. The values of K are illustrated in Table 3. These parameters were used to calculate R_L values; from Table 3, the R_L values were found to be 0.11 to 0.22, which identified the favorability of phosphate ion to be adsorbed onto BBA.

The Freundlich isotherm is another model that was applied in this investigation. The Freundlich constants K_f and n were calculated from the intercept and slope of the straight line, as shown in Figure 5. The values in the range of 2–10 represented good adsorption [28]. As shown in Table 3, the values were found to be in the range of 3.75–7.77. The Freundlich constant K_f was found to be related to adsorption capacity. The adsorption capacity at a temperature of 20 °C was found to be better than the other values obtained at other temperatures. Based on the findings of the isothermal models, BBA has a good affinity for P ions, and its adsorption capacity decreased with the increase or reduction in the optimal temperature (i.e., 20 °C).

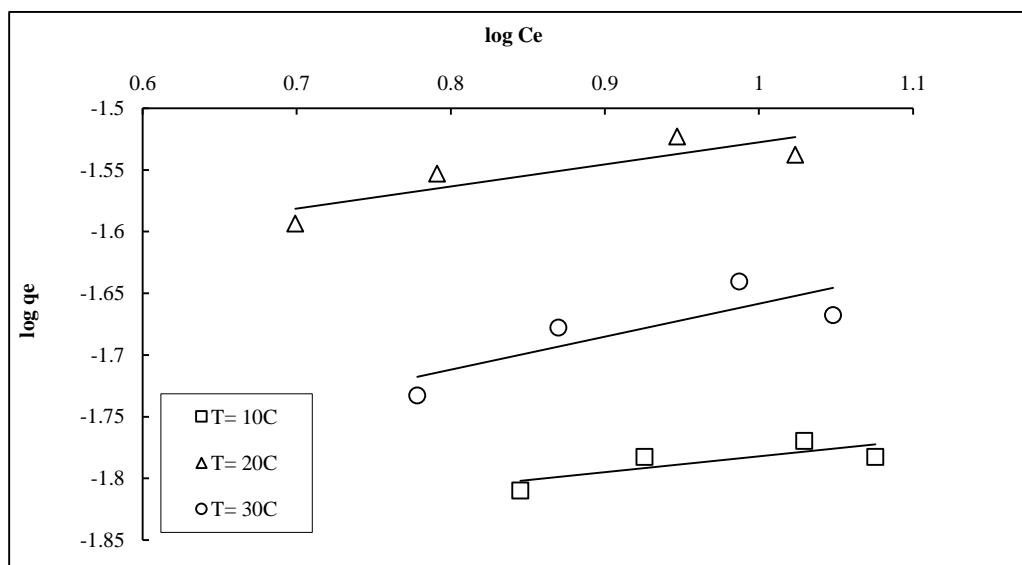


Figure 5. Freundlich isotherm model (log Ce vs. log qe) at initial pH 6.5, contact time 8 hr

It is true that ashes are chemically and physically different from each other, but most authors have indicated that the Langmuir model was the best isotherm model that explained the sorption via ashes [16, 30].

3.3. Effect of pH on Phosphate Removal

The adsorption of anions and cations at the liquid P–solid BBA interface seems to be influenced by the pH of the aqueous solution. The effect of variation of pH of the phosphate solution was measured by applying batch experiments at different pH (4, 5, 6, 7 and 8).

The results of the experiments, as shown in Figure 6, revealed that the phosphate removal efficiency of BBA was substantially dependent on the pH of the solution. It was apparent from the plot findings that the effectiveness of phosphate removal decreased with increasing pH of the solution from 4 to 8. After running the experiments for 8 h, the final phosphate concentrations were found to be 3.81 and 9.89 mg/L at pH 4 and 8, respectively. The experiments were

conducted at different contact times (1, 6 and 12 h) and a temperature of 20 °C. At all conditions, there was a decreased phosphate removal tendency of BBA with increasing pH of the solution. The highest removal efficiency of BBA was found at pH 4.

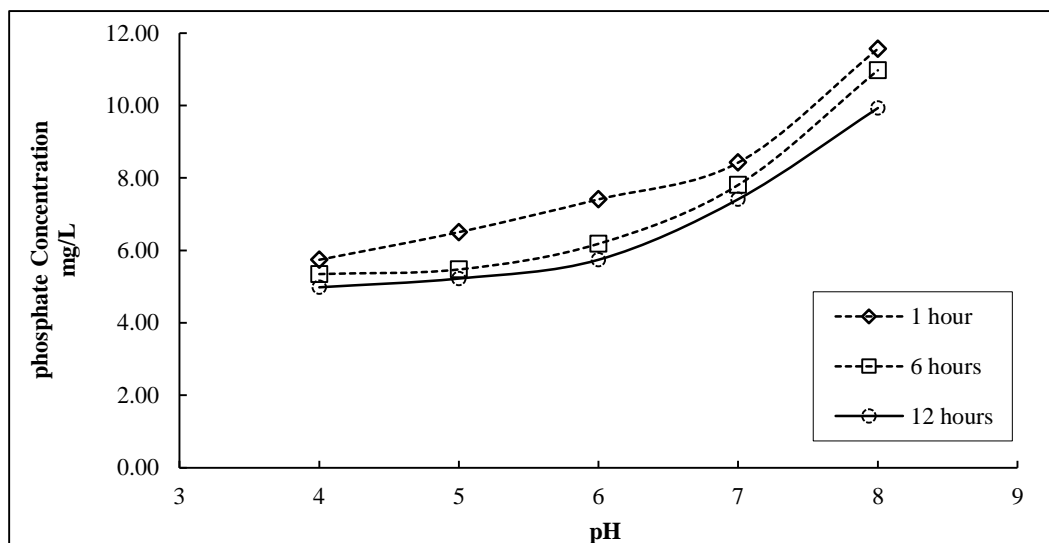


Figure 6. The effect of pH on the P removal

Multiple linear regressions were used as a mathematical tool to create a model for interpreting the relationship between the pH of the solution and phosphate sorption by BBA (qe). The pH and contact time between the phosphate solution and BBA were the independent variables in this model; while qe was the dependent variable. In addition, the exclusion of the contact time from the model was conducted as shown in model 2 of Table 4 to verify the influence of pH on phosphate sorption independently.

Table 4. Multiple linear regression models for phosphate sorption by furnace bottom ash

Model	Variables	Unstandardized Coefficients		R	R Square	Adjusted R Square	Std. Error of the Estimate
		B	Std. Error				
1	Constant	0.129	0.009	0.959	0.920	0.907	0.007284
	pH	-0.015	0.001				
	Time	0.002	0.000				
2	Constant	0.139	0.012	0.907	0.823	0.0809	0.010408
	pH	-0.015	0.002				

The equations 6 and 7 are obtained from the findings of multilinear regression models illustrated in Table 4.

$$q_{e_i} = 0.129 - 0.015 \text{ pH}_i + 0.002 T_i \tag{6}$$

$$q_{e_i} = 0.139 - 0.015 \text{ pH}_i \tag{7}$$

The value of phosphate sorption can be predicted from equation 6, but the validity of this model was proved at 20 °C. The standard error of the estimate in model 2 was 0.010408 and in model 1 was 0.007284. The little value of the standard error of the estimate showed a positive result, which refers to the improvement in the model depending on pH and time of contact. However, the models revealed that pH significantly influenced the phosphate sorption; the R-value did not decrease much when the contact time was excluded in model 2.

According to some studies [30, 31], the phosphate sorption capacity decreases with an increase in pH; the results of our study are also consistent with this finding. However, the studies did not find a mathematical model to link their findings of the pH to the phosphate sorption or other factors, as we did in this study.

4. Conclusion

The use of low cost adsorbent material for phosphate removal from wastewater is enhance the methods of treatment which is in line with the sustainability approach. BBA considered as a waste material and available for free. Therefore, using BBA as phosphate sorbent material is contributing in treatment waste by waste. The chemical composition of BBA indicated presence of oxides such as Fe, Al and Ca that might be the essential factors supporting the adsorption of

phosphate. The findings of this study revealed that the phosphate uptake onto BBA take place according to three phases (fast, moderate and slow). These phases bring our attention that removal process is issue of time more than it is issue of saturation. Specifically, if the majority of phosphate concentration take place in the fast phase as in case of BBA. Langmuir and Freundlich isotherm models are appropriate models for phosphate adsorption by BBA. It is evident through the values of RL, and n that both models are favorable for use However, the determination coefficient R^2 indicates that data fitted to Langmuir model is better than Freundlich model.

Furthermore, an increase in the pH of the solution caused an extreme decrease in the adsorption capacity. It might be due to the weakening of the electrostatic force of attraction between the negative charge of the phosphate and the positive charge of the BBA, which eventually caused the reduction of adsorption capacity. The key impact of this study is to enhance the research in utilizing the PSMs and use the method of this work as a strategy for examination the validation of the selected PSMs.

5. Conflicts of Interest

The authors declare no conflict of interest.

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