

# Phosphorus Removal from Aqueous Solutions by Bentonite: Effect of Al<sub>2</sub>O<sub>3</sub> Addition

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**ABSTRACT:** Phosphorus is the main nutrient responsible for eutrophication of aquatic ecosystems; therefore, it is important to develop new processes to remove phosphorus from aqueous solutions. In this research, performance, kinetics, and adsorbing isotherms of Bentonite and Bentonite- Alumina compound (10% Alumina (Al<sub>2</sub>O<sub>3</sub>), 90% Bentonite) were investigated. According to the results of this study, the Langmuir adsorption model was best to describe adsorption equilibrium data for Bentonite-Alumina mixture and Bentonite ( $r^2=0.999$  for Bentonite-Alumina mixture and  $r^2=0.996$  for Bentonite). The Langmuir Isotherm parameters for Bentonite and Bentonite-Alumina were  $Q_m=15.2, 18.1$  mg/g and  $K=4.3, 5.5$  L/mg, respectively. Moreover, the Freundlich Isotherm parameters were  $K=6.6, 9.12$  and  $n=3.05, 3.7$ , respectively. Bentonite-Alumina mixture was found to more effective for the removal of phosphorus than Bentonite (97% in 140 min in comparison to 90% in 140 min).

**Keywords:** Phosphorous Removal, Absorbents, Bentonite, Alumina, Algae, Eutrophication

ORIGINAL ARTICLE

## INTRODUCTION

The generation of residual sludge may remain unavoidable with current water treatment technologies. Aluminium based water treatment sludge is generated at those water treatment plants where aluminium sulphate is used as the primary coagulant together with organic polymer as co-coagulant. Such sludge contains removed colour, turbidity and humic substances from sources water, plus sludge aluminium sulphate and residual polymers (Dayton and Basta, 2007). On the other hand, the targets of phosphorus control set by phosphorus regulation (1998) of the Irish environmental protection agency remain a very considerable challenge for the vast majority of local authorities although the monitoring indicated that the majority of Irelands Rivers and lakes are not polluted. There is evidence to suggest that problems remain in a number of rivers as a result of municipal pollution and ineffective wastewater treatment especially phosphorus removal facilities (Clenaghan et al. 2005). Drinking-water treatment sludge (DWTS) is a by-product generated during the production of drinking water where aluminum or iron based salts are used as coagulants to remove color, turbidity and humid substances. Current legislation classifies DWTS as waste and usually it is chemically conditioned and mechanically dewatered before disposal in landfills. Various alternative options for disposal, regeneration and beneficial reuse of DWTS have been explored in the past decade (Babatunde and Zhao, 2007). Aluminum or iron hydroxides are the important component of the sludge once it has been dewatered, this opens the possibility of reusing it to control pollutant such as phosphorus since aluminum or

iron can enhance their adsorption and chemical precipitation. Extensive studies have shown the effectiveness of DWTS for phosphorus immobilization in soils (Makris et al., 2005). Phosphorus (P) is an essential, often limiting, nutrient for growth of organisms in most ecosystems. However, excessive supply of phosphorus from wastewater into water bodies, such as lakes, rivers and creeks cause eutrophication, resulting in the bloom of aquatic plants, growth of algae and depletion of dissolved oxygen. Phosphorus removal from wastewater has been widely studied during the past decades. Typical removal methods such as chemical and biological treatments have been successfully applied (Yeoman et al., 1998). Nevertheless, increasing attention has been paid to adsorptive removal of phosphate from aqueous solution (Hano et al., 1997; Donnert, 1999). Eutrophication has become the primary water quality issue for most of the freshwater and coastal marine ecosystems in the world. It is one of the most visible examples of human changes to the biosphere, affecting aquatic ecosystems from the Arctic to the Antarctic (Smith and Schindler, 2009; Smith and Schindler, 2006). More than 40% of water bodies in many regions of the world are considered to have eutrophication problems. One gram of phosphorous is required for every seven grams of nitrogen for the formation of the organic matter created in the process (Ross et al., 2008). Thus, the excess of bioavailable phosphorous is the key nutrient which is understood to lead to eutrophication of water bodies, resulting in increased aquatic plant and algal growth. A recent study of over 14,000 lakes <1(ha) in size in Great Britain found that 51% are likely to require p- reduction measures to meet the water framework directive to achieve 'good

**Table. 1.** Literature review on adsorptive media for phosphorus removal [12]

Aluminum	Calcium	Iron	Industrial by-products	Clay
1- Activated alumina (7gP/kg) 2- Shale (0.73gP/kg) 3- Bauxite (0.36gP/kg) 4- Zeolite (2.15gP/kg)	1- <u>Wollastonite</u> (0.85gP/kg) 2- Limestone (0.68gP/kg) 3- Dolomite (0.30gP/kg)	1- Iron oxide (12.65gP/kg) 2- Iron coated sand (1.50gP/kg) 3- Red mud (0.58gP/kg)	1- Steel slag (18gP/kg) 2- Blast furnace slag (8.89gP/kg) 3- Coal fly ash (8.81gP/kg)	1- <u>Allophane</u> ( $q=5.620C^{0.264}$ ) 2- <u>Phoslock</u> (11-12gP/kg) 3- <u>Bentonite</u> (0.42mgP/kg)

Chemical methods for removing phosphorus compounds caused product quality reducing and side effects on aquatic environments. Among the available methods for removing these pollutants, adsorption is still one of the most preferred methods, especially for effluents with moderate and low pollutant concentration (Douglas and Coad, 1996; Nameni et al., 2007). In the past years, there has been increasing interest in developing recyclable inorganic adsorbents, particularly from clay for efficient removal of organic pollutants from aqueous solutions (Dable et al., 2008; Nouri, 2002). In this study, the performance of Bentonite-Alumina mixture (10% Alumina, 90% Bentonite) in removing phosphorus from aqueous solutions was investigated. The adsorption kinetics, adsorption behavior and adsorption isotherms were studied. Langmuir and Freundlich adsorption models were used in this research. Bentonite is an absorbent aluminum phyllosilicate, essentially impure clay consisting mostly of montmorillonite. The absorbent clay was given the name bentonite by Wilbur C. Knight in 1898, after the Cretaceous Benton Shale near Rock River, Wyoming. There are different types of bentonite, each named after the respective dominant element, such as potassium (K), sodium

(Na), calcium (Ca), and aluminium (Al). Experts debate a number of nomenclatorial problems with the classification of bentonite clays. Bentonite usually forms from weathering of volcanic ash, most often in the presence of water. However, the term bentonite, as well as similar clay called tonstein, has been used to describe clay beds of uncertain origin. For industrial purposes, two main classes of bentonite exist: sodium and calcium bentonite. In stratigraphy and tephrochronology, completely devitrified (weathered volcanic glass) ash-fall beds are commonly referred to as K-bentonites when the dominant clay species is illite. Other common clay species, and sometimes dominant, are montmorillonite and kaolinite. Kaolinite-dominated clays are commonly referred to as tonsteins and are typically associated with coal.

### Adsorption isotherms

Langmuir isotherm (Langmuir, 1918) is most widely used for the sorption of a pollutant from a liquid solution (Del Bubba et al., 2003; Allen et al., 2003; Ho, 2004). The model assumes that the sorption takes place at specific homogenous sites within the adsorbent, i.e. once

a sorbate molecule occupies a site; further adsorption at this site is impossible (Linderfelt et al., 1997). The equation of Langmuir is represented as follows:

$$q_e = \frac{Q_0 b C_e}{1 + b C_e} \quad (1)$$

Where  $q_e$  the mass of P is adsorbed on adsorbent at equilibrium (mg/g);  $C_e$  is the equilibrium concentration of P solution (mg/l);  $Q_0$  is the maximum adsorption capacity (mg/g);  $b$  is a Langmuir constant (l/mg) related to the energy of adsorption. Physically,  $b$  is a measure of the affinity of the adsorbate for the adsorbent. The linear form of the Langmuir isotherm is:

$$\frac{C_e}{q_e} = \frac{C_e}{Q_0} + \frac{1}{b Q_0} \quad (2)$$

Hence a plot of  $\frac{C_e}{q_e}$  versus  $C_e$  or  $\frac{1}{q_e}$  versus  $\frac{1}{C_e}$  gives a straight line to determine the isotherm parameters.

### Freundlich isotherm

Although Freundlich isotherm (Freundlich, 1906) is an empirical equation to describe heterogeneous adsorption systems, it is the earliest known isotherm (Ho, 2004). This isotherm is given as:

$$q_e = K_F C_e^{\frac{1}{n}} \quad (3)$$

A linear form of the Freundlich expression is:

$$\text{Log } q_e = \text{Log } K_F + \frac{1}{n} \text{Log } C_e \quad (4)$$

where  $K_F$  is the Freundlich constant (l/g) related to the bonding energy; and  $1/n$  is the heterogeneity factor in which  $n$  is a measure of the deviation from linearity of the adsorption. The value of  $n$  indicates the degree of non-linearity between P solution concentration and adsorption as follows: if the value of  $n$  is equal to unity, the adsorption is linear; if the value is below to unity, this implies that adsorption process is chemical; and if the value is above to unity, adsorption is a favorable physical process (Kundu and Gupta, 2006). The plot of  $\text{Log } q_e$  vs  $\text{Log } C_e$  was employed to generate the intercept value of  $K_F$ . and the slope of  $1/n$ .

### Temkin isotherm

The Temkin isotherm has been used in the following form (Choy et al., 1999; Ozacar, 2003):

$$q_e = \frac{RT}{b_T} \ln(AC_e) \quad (5)$$

A linear form of the Temkin isotherm can be expressed as:

$$q_e = \frac{RT}{b_T} \ln(A) + \frac{RT}{b_T} \ln(C_e) \quad (6)$$

Where  $\frac{RT}{b_T} = B$ ; R is the gas constant (8.31J/Mol K) and T is the absolute temperature. The sorption data can be analyzed according to Eq. (6). Therefore a plot of  $q_e$  versus  $\ln(C_e)$  enables one to determine the constants A and  $b_T$ . Temkin isotherm is based on the assumption that heat of adsorption would decrease linearly with increase of coverage of adsorbent due to sorbate/sorbent interaction.

### Dubinin-Radushkevich isotherm

The Dubinin-Radushkevich (Dubinin, 1960) isotherm assumes that there is a surface area where the adsorption energy is homogeneous. The Dubinin-Radushkevich equation has the following form:

$$q_e = q_m \cdot e^{-\beta \varepsilon^2} \quad (7)$$

A linear form of Dubinin-Radushkevich isotherm is:

$$\ln q_e = \ln q_m - \beta \varepsilon^2 \quad (8)$$

where  $q_m$  is the Dubinin-Radushkevich monolayer capacity (mg/g),  $\beta$  a constant related to sorption energy, and  $\varepsilon$  is the Polanyi potential which is related to the equilibrium concentration as follows:

$$\varepsilon = RT \ln \left( 1 + \frac{1}{C_e} \right) \quad (9)$$

The constant  $\beta$  gives the mean free energy,  $E$ , of sorption per molecule of the sorbate when it is transferred to the surface of the solid from infinity in the solution and can be computed using the relationship (Choy et al., 1999; Lin and Juang, 2002):

$$E = \frac{1}{\sqrt{2\beta}} \quad (10)$$

### Error Analysis

For the purpose of quantitatively comparing the applicability of different adsorption isotherms in fitting to data, the following four error functions were employed for error analysis:

1. The sum of the squares of the errors (SSE) (Kundu and Gupta, 2006):

$$SSE = \sum_{i=1}^n (q_{e,cal} - q_{e,exp})^2 \quad (11)$$

2. The sum of the absolute errors (SAE) (Kundu and Gupta, 2006):

$$SAE = \sum_{i=1}^n |q_{e,cal} - q_{e,exp}| \quad (12)$$

3. The hybrid fractional error function (HYBRID) (Kundu and Gupta, 2006):

$$HYBRID = \frac{100}{n-p} \sum_{i=1}^n \left[ \frac{(q_{e,exp} - q_{e,cal})^2}{q_{e,exp}} \right]_i \quad (13)$$

4. Normalized percentage standard deviation (NPSD) (Sun and Yang, 2003):

$$NPSD(\%) = 100 \sqrt{\frac{1}{n-1} \sum_{i=1}^n \left( \frac{q_{e,exp} - q_{e,cal}}{q_{e,exp}} \right)^2} \quad (14)$$

where  $q_{e,exp}$  and  $q_{e,cal}$  are respectively the experimental values and calculated values by adsorption isotherm;  $n$  and  $p$  refer to the number of data points and the number of isotherm parameter, respectively.

## MATERIALS AND METHODS

The Bentonite (clay) was used in dust form in this study. The chemical composition of the Bentonite was obtained through XRF and wet chemical analysis is presented in Table 2. The CEC for Bentonite was 100.19 meq/100gr (NH4AC method) and the percent concentration of monmorillonite in Bentonite was 75.92%.

**Table 2.** Chemical analysis of Bentonite

Bentonite	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	LOI	Total
	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	
	53.72	19.12	0.85	4.93	3.29	5.28	3.46	0.44	8.75	100

### Stock phosphorus solution

A stock phosphorus solution with a concentration of 40 mg/L was prepared by dissolving 175.75 mg KH<sub>2</sub>PO<sub>4</sub> in 1 Lit of pure water and used to prepare solutions with lower concentrations.

### Experiments

Experiments were carried out in two phases; first phase was for obtaining the phosphorus removal efficiency for two adsorbents Bentonite and Bentonite–Alumina. Second phase of experiments was for obtaining the adsorption capacity of the two adsorbents. In the first phase, 1g of Bentonite as an adsorbent was disposed in 14 250<sup>cc</sup> beakers and then 100<sup>cc</sup> of phosphorus solution with concentration of 8mg/L was added to each beaker. Beakers were mixed at 100 rpm for 1 min and then placed in ambient temperature until settled. Samples phosphorus concentrations were measured regularly by spectrophotometry method. Phosphorus removal efficiency ( $\eta$ ) was calculated according to the equation below:

$$\eta = \frac{C_0 - C_e}{C_0} \times 100 \quad (15)$$

where  $C_0$  and  $C_e$  are initial and equilibrium phosphorus concentrations in solution (mg/Lit).

In the second phase of experiments, for determining of adsorption capacity, 1mg of selected adsorbents and 10 mL of phosphorus solution with different concentrations (5, 4, 3, 2.5, 2, 1.5, 1, and 0.5 mg/L) were prepared. These solutions were then mixed at 100 rpm for 10 min and placed in ambient temperature until settled. Samples were filtered with Whatman (0.45  $\mu$ m) and then the phosphorus concentrations of filtered samples were measured by spectrophotometry method. Adsorption capacity (Q) was calculated according to the equation below:

$$Q = \frac{C_0 - C_e}{m} \times V \quad (16)$$

Where  $C_0$  and  $C_e$  are initial and equilibrium concentration (mg/L),  $V$  is the solution volume (mL) and  $m$  is the amount of absorbents (g). All the experiments were carried out according to standard method 20<sup>th</sup> edition (standard method for examination of water and wastewater, 1992).

## RESULTS AND DISCUSSION

Figures 1 and 2 represent the phosphorus concentration and phosphorus removal for two absorbents (Bentonite and Bentonite-Alumina). According to the results, phosphorus removal was higher in the presence of  $Al_2O_3$ . A decrease of 97 percent in phosphorus concentration was observed for Bentonite-Alumina mixture during the maximum contact time 140 min as compared to 90% for Bentonite. Although the initially kinetics of phosphorus removal was faster for Bentonite, phosphorus concentration remaining in solution after 100 min (equilibrium time) was below 0.2 mg/L for Bentonite-Alumina in comparison to Bentonite. Replacing 10 percent Bentonite with  $Al_2O_3$  seems to indicate that adsorption strength was improved significantly because of probably increasing the adsorption surface area and polarity of Bentonite due to alumina addition.

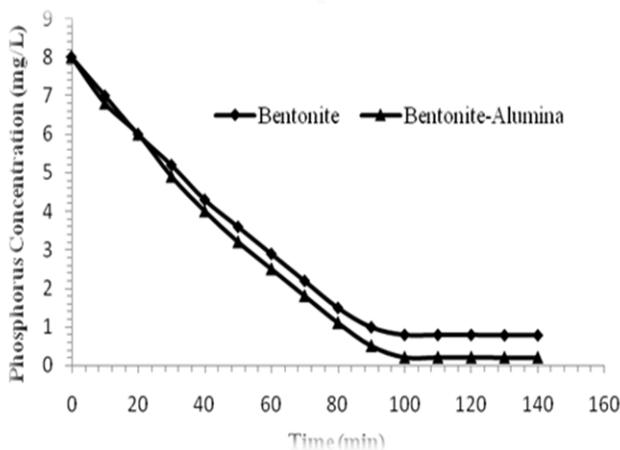


Figure 1. Phosphorus Concentration in solution

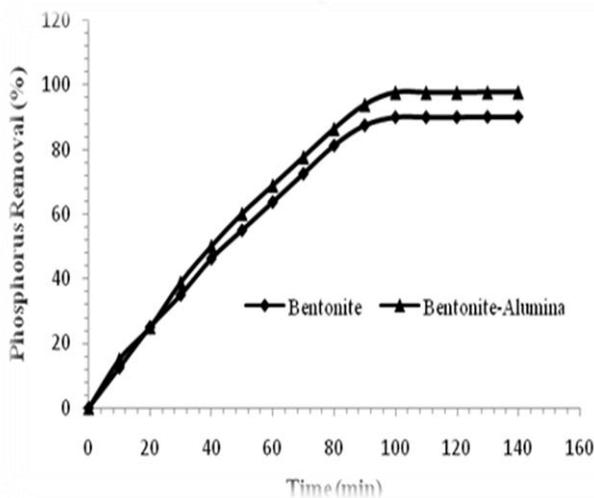


Figure 2. Phosphorus removal efficiency

The adsorption capacity of Bentonite was improved by replacing with 10 percent  $Al_2O_3$ . Adsorption capacity of Bentonite-Alumina was about 18mg/g in equilibrium concentration of 3.6 mg/L as compared to 14 mg/g for Bentonite. This result might be attributed to increasing the adsorption surface area and polarity of Bentonite by replacing with 10%  $Al_2O_3$ .

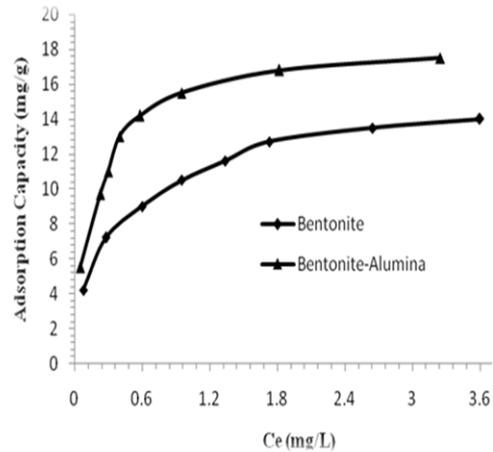


Figure 3. Adsorption Capacity of two absorbents

### Adsorption Isotherms

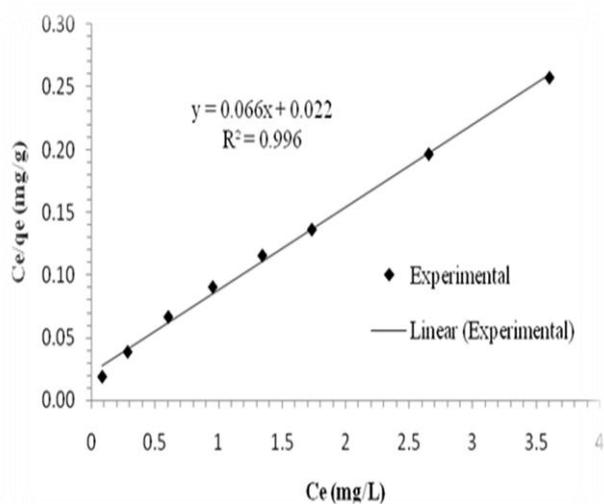
Mention Figure 3 in the text Different isotherm models such as Langmuir and Freundlich were used to describe ions distribution Explain why the other models discussed earlier – Temkin and DR – were not used. In order to find most appropriate model for the phosphorus adsorption, the data were fitted to each isotherm model. The obtained isotherm parameters and correlation coefficients ( $R^2$ ) are presented in Table 4. Figures 4 and 7 represent the correlation coefficients of Langmuir and Freundlich adsorption models for the two absorbents. According to the results, the Langmuir adsorption model was best to describe adsorption equilibrium data for Bentonite-Alumina mixture and Bentonite ( $r^2=0.999$  for Bentonite-Alumina mixture and  $r^2=0.996$  for Bentonite) as compared to Freundlich adsorption model. Correlation coefficients ( $r^2$ ) of the Freundlich model for Bentonite and Bentonite-Alumina were 0.977 and 0.916, respectively.

Table 3. Isotherm equations

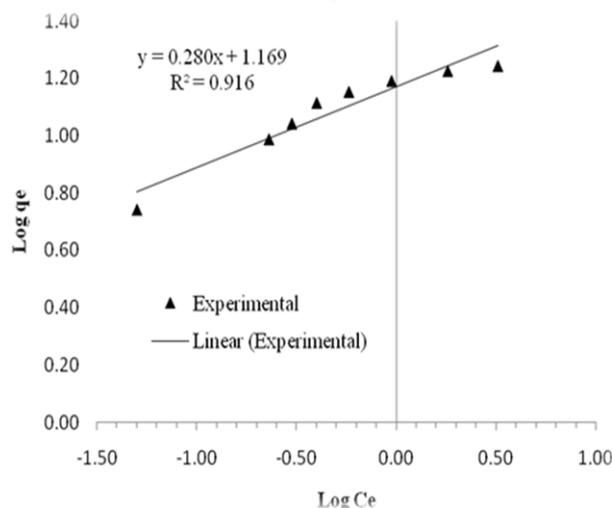
Isotherm name	Isotherm equation	Parameters
Langmuir	$q_t = \frac{Q_m K C_e}{1 + K C_e}$	$C_e$ : the equilibrium concentration (mg/L) $q_t$ : the amount adsorbed per amount of adsorbent at the equilibrium (mg/g) $Q_m$ (mg/g) and $K$ (L/mg): the Langmuir constants related to the maximum sorption capacity and energy of adsorption, respectively.
Freundlich	$q_t = K C_e^{\frac{1}{n}}$	$K$ (mg/g): an indicator of the adsorption capacity $\frac{1}{n}$ (mg/L): adsorption intensity

Table 4. Adsorption isotherm constants

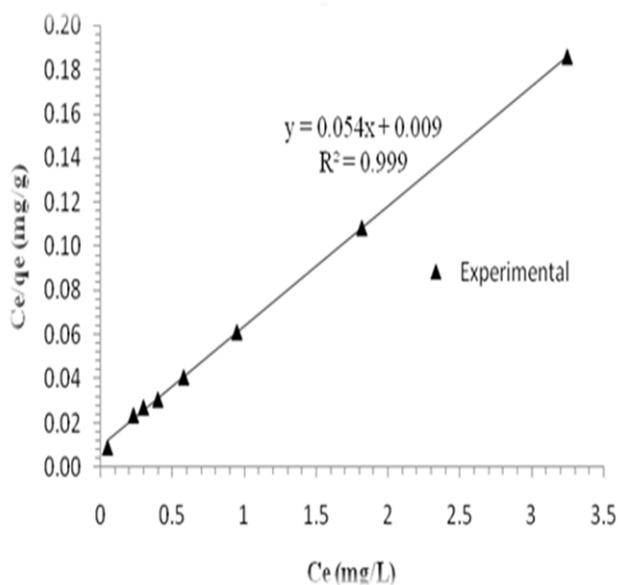
Isotherm type	Isotherm constants		$R^2$	
	Bentonite	Bentonite-Alumina	Bentonite	Bentonite-Alumina
Langmuir	$Q_m=15.2$ mg/g $K=4.3$ L/mg	$Q_m=18.1$ mg/g $K=5.5$ L/mg	0.996	0.999
Freundlich	$K=6.6$ mg/g $n=3.05$ L/mg	$K=9.12$ mg/g $n=3.70$ L/mg	0.977	0.916



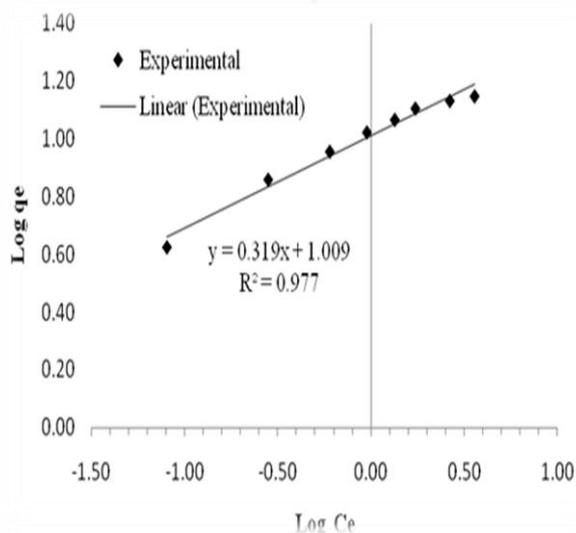
**Figure 4.** Langmuir isotherm (Bentonite, Bentonite-Alumina) - (a). Bentonite



**Figure 7.** Freundlich isotherm (Bentonite, Bentonite-Alumina)- (b). Bentonite-Alumina



**Figure 5.** Langmuir isotherm (Bentonite, Bentonite-Alumina)- (b). Bentonite-Alumina



**Figure 6.** Freundlich isotherm (Bentonite, Bentonite-Alumina)- (a). Bentonite

## CONCLUSION

In the present study, the ability of Bentonite and Bentonite-Alumina to remove phosphorus from aqueous solutions has been investigated in maximum contact time of 140 min. Bentonite-Alumina was found to be more effective for the removal of phosphorus than Bentonite. The removal of phosphorus using Bentonite-Alumina was reached 97% at ambient temperature, equilibrium time (120 min) and mild agitation (100 rpm) as compared to 90% for Bentonite. Adsorption isotherms have been determined and data have been analyzed according to Langmuir and Freundlich models. Langmuir adsorption model was found to be best to describe adsorption equilibrium data for Bentonite ( $r^2=0.996$ ) and Bentonite-Alumina ( $r^2=0.999$ ). Finally, this study showed that Bentonite-Alumina could be regarded as one of the efficient adsorbent of phosphorus from aqueous solutions.

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