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Phosphate removal from wastewater by using water treatment sludge

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ABSTRACT

In this study, the potential of alum sludge, obtained from a drinking water treatment plant located in Istanbul, Turkey, as an adsorbent for the removal of phosphate from aqueous solution was investigated. The effects of adsorbent amount, pH, initial concentration, and temperature on phosphate removal were studied by batch adsorption experiments. Equilibrium data were analyzed according to the Langmuir and the Freundlich isotherms. The Freundlich isotherm equation provided a better fit than the Langmuir isotherm equation. Thermodynamic parameters (ΔG, ΔH, and ΔS) related to the process were also calculated and reported. The positive value of ΔH and ΔS indicated the endothermic nature of the phosphate adsorption and the affinity of the adsorbent for the phosphate, respectively.

Keywords: Adsorption; Isotherm; Phosphate removal; Thermodynamic; Water treatment plant sludge

1. Introduction

In conventional water treatment plants, drinking water is obtained from raw water via a series of processes, which are coagulation, flocculation, sedimentation, and filtration. In the process of coagulation, Fe or Al salts are used. These salts form flocs with impurities in raw water. Following the sedimentation process, partially clarified water is filtered out. Such a water treatment plant generates a solid residue (alum or ferric sludge) with high water content during water treatment. The produced water treatment sludges (alum or ferric sludge) should be well treated and disposed of so that they do not cause damage to the environment. The sludges are dewatered before disposal. Therefore, a cake with a concentration of solids of 60% to 70% is obtained. The dewatered sludge cake can be used as fertilizer, incinerated, disposed at landfills, or composted with urban waste [1]. As stated by Ahmad et al. [2], the reuse of water treatment plant sludges is a favorable and sustainable solution for the final disposal of these wastes.

The water treatment plant sludges mainly contain clay minerals, very fine-grained minerals (mainly oxides and hydroxides of aluminum and iron), organic matter and contaminants from the wastewater discharges [1]. The chemical composition of the sludge is important for the selection of the potential reuse alternatives. However, to provide safe and sustainable disposal for the sludges, in addition to the chemical properties, the physical properties should also be taken into account [2].

The physical and chemical compositions of alum sludge vary depending on the treatment method and the treatment plant [3]. Besides, the chemical composition of the sludge depends on the raw water quality [2–4], the nature of the chemicals used [2,3], the origin of the raw water (surface water or groundwater), the soil type of the region, the chemical products present [1], and the dosage applied [4].

Up till now, various research studies were carried out on the reuse of alternatives of alum sludges from drinking water treatment plants in wastewater treatment. Alum sludge was examined for its ability to remove various pollutants such as

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phosphate [5–7] from aqueous media. Alum sludges, which were obtained from local water treatment plants in different countries, were found to be good adsorbents for the removal of phosphate from wastewater.

In this study, phosphate removal from aqueous solution using alum sludge collected from a drinking water treatment plant in Istanbul, Turkey was investigated. The effects of operating parameters, including alum sludge dosage, initial pH and temperature on phosphate adsorption were examined. As far as we know from the literature [5,6,8–10], up to now thermodynamics of phosphate adsorption on alum sludge were not analyzed and discussed in the previous studies. In this study, in addition to the adsorption equilibrium, thermodynamics was also analyzed and discussed.

2. Materials and methods

2.1. Adsorbent

Dewatered alum sludges were obtained from the Kağıthane Drinking Water Treatment Plant of Istanbul Water and Sewerage Administration (ISKI), Turkey. They were heated in an oven at 105°C to obtain constant weight. The oven-dried material was ground with a pestle and mortar, and then sieved to size 0.25–0.5 mm particles. Fig. 1 shows oven-dried and sieved alum sludge.

Some physical characteristics of the alum sludge used in this study were analyzed in the TÜBİTAK Marmara Research Center in Gebze Kocaeli, Turkey.

2.2. Adsorbate

Phosphate stock solution was prepared by dissolving KH_2PO_4 in deionized water. The stock solutions were further diluted with deionized water to form various phosphate concentrations.

2.3. Adsorption studies

Batch adsorption studies were carried out in 250 mL Erlenmeyer flasks. The flasks containing the desired amount of adsorbent and 100 mL test solutions were shaken in an incubator shaker (Daihan WIS-10R) at 150 rpm. Different adsorbent amounts (0.25, 0.50, 1.0, 1.5, 2, 2.5, 3, 3.5, 4, 4.5, 5, 5.5, 6, and 6.5 g per 100 mL), initial pH (2, 3, 4, 5, 6, 7, and 8), and temperature (20°C, 30°C, 40°C, and 50°C) were assessed. Isotherm studies were conducted with various initial phosphate concentrations (C_0: 2.5–50 mg/L) at five different temperatures (10°C, 20°C, 30°C, 40°C, and 50°C). At the end of each experiment, the flask contents were centrifuged at 35,000 rpm for 10 min (Hettich EBA 20) to separate solids and liquids. The supernatants were analyzed for phosphate (PO_4^{3-}–P) concentration using ammonium molybdate spectrophotometric method according to the procedure stated in the Standard Methods [11] with a UV/VIS spectrophotometer (Split Beam PGI T60). pHs of the solution was measured by a pH meter (Orion Dual Star), and pH adjustments were carried out with 0.1 M HCl or NaOH solution, when necessary.

The percent removal ($R(\%)$) of phosphate is defined as:

$$R(\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \tag{1}$$

The adsorption capacity (q) was calculated from the following equation:

$$q = \frac{(C_0 - C_e) \times V}{m} \tag{2}$$

where C_0 and C_e are the initial and equilibrium concentrations of phosphate in the solution (mg/L), respectively, V is the volume of phosphate solution (L), and m is the amount of adsorbent used (g).

Experiments were conducted in duplicate at least, and the mean values were presented.

2.4. Theory

2.4.1. Equilibrium isotherms

Langmuir model (Eq. (3)) [12] and Freundlich model (Eq. (4)) [13] are expressed in linear form as follows:

$$\frac{C_e}{q_e} = \frac{1}{bq_{\max}} + \frac{C_e}{q_{\max}} \tag{3}$$

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \tag{4}$$

where C_e is the equilibrium phosphate concentration (mg/L), q_e is the amount of phosphate uptake per unit weight of the adsorbent (mg/g), $q_{\max}$ is the maximum adsorption capacity of the adsorbent (mg/g), b is the Langmuir isotherm constant (L/mg), and K_F (mg/g) and $1/n$ are the Freundlich constants representing the adsorption capacity and adsorption intensity, respectively.

2.4.2. Thermodynamics

The thermodynamic parameters such as the change in free energy (ΔG), enthalpy (ΔH) and entropy (ΔS) were calculated using the following equations:

$$\Delta G = -RT \ln K_d \tag{5}$$

$$\ln K_d = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \tag{6}$$

where K_d is the distribution coefficient for the adsorption. ΔH, ΔS, and ΔG are the changes in enthalpy, entropy, and Gibb's free energy, respectively, R is the gas constant, and T is the absolute temperature.

3. Results and discussion

3.1. Characterization of the adsorbent

Some physical characteristics of the alum sludge used in this study are given in Table 1. The specific surface area of the alum sludge was found to be 91.77 m^2/g. The surface area of the alum sludge taken from a water treatment plant in Dublin, Irland was reported as 49.03 m^2/g by Yang et al. [10].

Abo-El-Enein et al. [14] stated that the surface areas of the alum sludges collected from Al-Fustat drinking water treatment plant, Egypt, and thermally treated at 100°C, 500°C, and 700°C were found to be 22.00, 83.10, and 68.27 m^2/g, respectively. Awab et al. [15] revealed that the alum sludge was taken from the Semanggar Water Treatment Plant, Kota Tinggi, Johore, Malaysia had a surface area value of 38.92 ± 2.68 m^2/g. The local alum sludge (Istanbul, Turkey) investigated in this study was found to have a higher surface area than the aforementioned local alum sludges and showed a type IV adsorption isotherm characteristic of mesoporous solids (Fig. 2) [16].

As can be seen from Table 1, the average pore diameter of the alum sludge used in this study is 68.9 Å, which indicates that the alum sludge consists of mesopores [17]. As a comparison, Abo-El-Enein et al. [14] reported that the average pore diameters (mean pore radius) of the alum sludges collected from Al-Fustat drinking water treatment plant, Egypt, and thermally treated at 100°C, 500°C, and 700°C was found to be 57.95, 50.01, and 69.15 Å, respectively. The pore size distribution of the local alum sludge used in this study is shown in Fig. 3.

3.2. Effect of amount of adsorbent

The effects of the amount of adsorbent on adsorption capacity, q (mg/g), and phosphate removal (%) are shown in Fig. 4. It was observed that increasing the amount of alum sludge increased phosphate removal (%). The percentage removal of phosphate continued to increase slowly beyond about 20 g/L and almost reached a plateau value. This phenomenon can be described as stated by Mor et al. [18]; while the increase in phosphate removal (%) in high amount of adsorbent may be due to more free sites causing increase in the net surface area of the adsorbent, the slow increase in phosphate removal (%) in further increase in the amount of the adsorbent may be due to the aggregation of the adsorbent.

Fig. 1. Oven-dried and sieved alum sludge.

Table 1
Physical properties of the local alum sludge in Istanbul, Turkey

Properties	Values
Surface area (multipoint Brunauer–Emmett–Teller (BET))	91.77 m^2/g
Average pore diameter	68.9 Å
Total pore diameter[a]	0.16 cc/g
Pore volume	1.151 cc/g
Half pore width	0.003 μ

[a] Total pore diameter for pores with diameter less than 201.00 nm at P/P_0 = 0.990368.

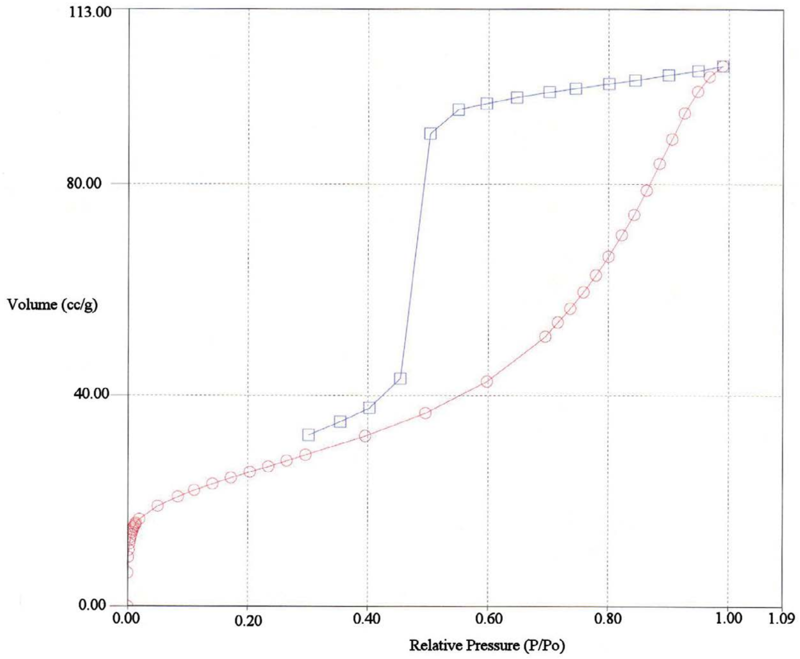

Fig. 2. Nitrogen adsorption isotherms of the local alum sludge (○: adsorption, □: desorption).

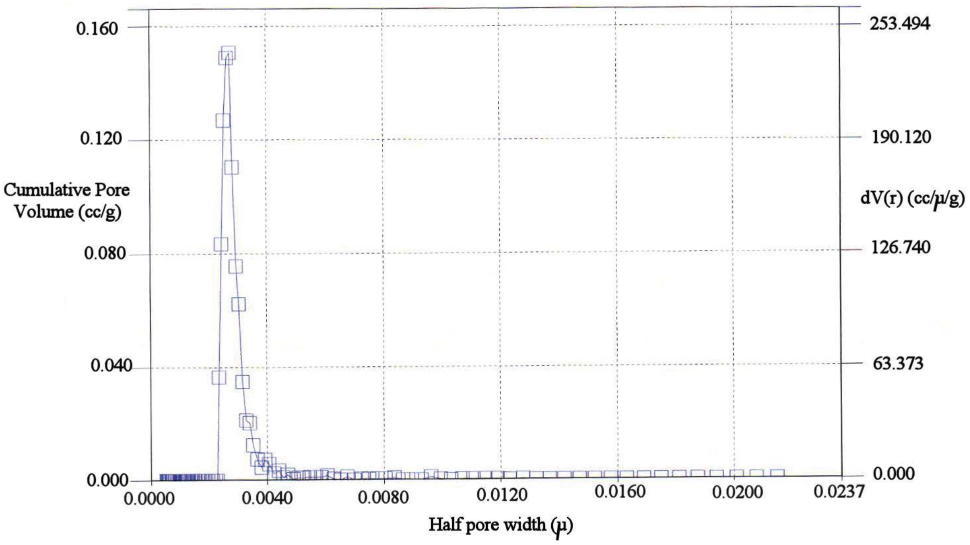

Fig. 3. Pore size distribution of the local alum sludge.

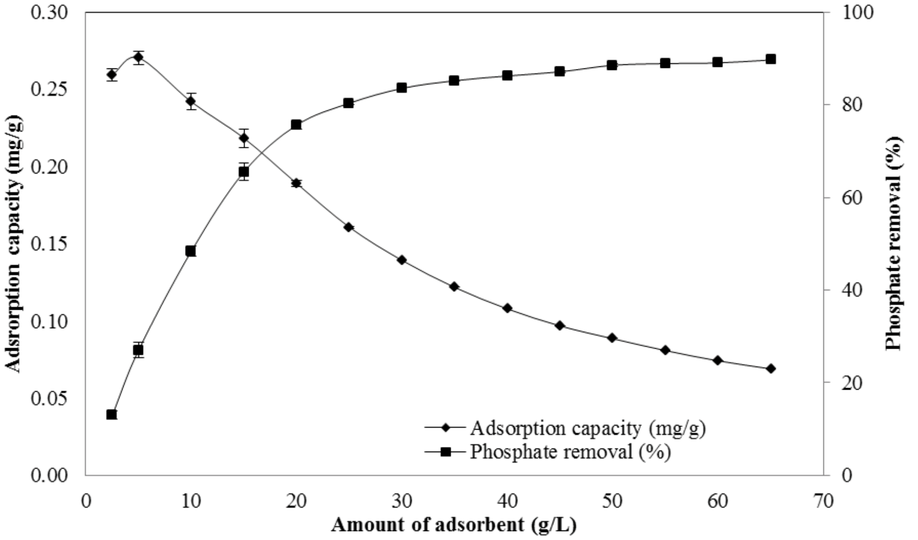

Fig. 4. Effect of the amount of adsorbent on adsorption; (a) adsorption capacity, *q* (mg/g) and (b) phosphate removal (%) (C_0: 5 mg/L, T: 25°C, *t*: 60 min).

As can be seen from Fig. 4, adsorption capacity (mg/g) decreases with the increasing amount of alum sludge. A similar trend was observed in the literature for various adsorbents such as agro-waste rice husk ash [18], zirconium-modified zeolite [19], crawfish char derived at different pyrolysis temperature [20], and hierarchical CuAl/biomass carbon fiber layered double hydroxide [21].

3.3. *Effect of pH on adsorption*

The effect of pH on the adsorption capacity of alum sludge for phosphate is shown in Fig. 5. As can be seen from the figure, the adsorption capacities of phosphate on alum sludge increased gradually with the increase of pH from 2 to 6. However, the adsorption capacity of alum sludge for phosphate decreased for pHs above 6. A review of the literature shows that the highest removal efficiency for phosphate by alum sludge was achieved at pH 5–6 [7,22].

As described by Chubar et al. [23], phosphate exists in solution as H_3PO_4, $H_2PO_4^-$, HPO_4^{2-}, and PO_4^{3-} at different ratios according to pH. At pH values 3–5, one-charged species of phosphate ($H_2PO_4^-$) is dominating, which is the most easily absorbed form of phosphate. Decrease of adsorption capacity with the increase of pHs above 6 may be attributed to the increase of the portion of one-charged phosphate species that have less affinity to the surface. Besides, it can be said that since the surface of the alum sludge has a more negative surface charge above pH 6, electrostatic repulsive interaction between negatively charged alum sludge surface and phosphate ions occurs. Similar findings were reported for phosphate removal by adsorbents such as crawfish char [20], hierarchical CuAl/biomass carbon fiber layered

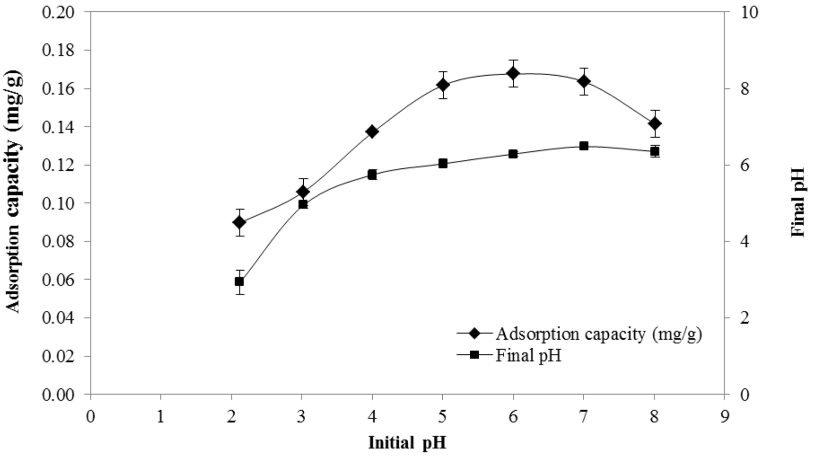

Fig. 5. Effect of pH on adsorption of phosphate ions on alum sludge (a) adsorption capacity and (b) final pH (C_0: 5 mg/L, *m*: 25 g/L, *t*: 30 min, *T*: 25°C)

double hydroxide [21], novel inorganic ion exchangers [23], Mg-laden biochar [24], and raw peat and peat modified with iron(III) hydroxy ions [25].

In this study, the change of pH values of the solution was traced during the adsorption of phosphate by alum sludge. The phosphate adsorption at the initial pH range of 2–8 exhibits that the final pHs are higher than those before adsorption. The final pH increased from 2.71 to 6.45 with increasing pH till pH 7 and decreased to 6.24 in pH 8. This phenomenon could be due to the OH^- from alum sludge surface releasing into solution, which leads to the protonation of the adsorbent. As stated by Jiang et al. [24], the protonated adsorbent surface was preferred for phosphate adsorption. At pH 8, the final pH was lower than that before adsorption. It may be due to the release of H^+ from the adsorbent's surface into the solution, which reduces the phosphate adsorption owing to the deprotonation of the adsorbent.

Yang et al. [5] studied the mechanisms of phosphate adsorption on dewatered alum sludge. They revealed that the surface of the alum sludge contains a considerable amount of reactive functional groups (–OH, –Cl, $-SO_4$ and humic substances) which were responsible for the ligand exchange mechanism of the phosphate adsorption. Phosphate replaces the functional groups from the surface of the alum sludge. They also stated that the other mechanisms contributing to the phosphate adsorption were chemical reaction and precipitation.

3.4. Adsorption isotherms

The experimental adsorption data for phosphate onto the alum sludge, which was obtained at four different temperatures (from 10°C to 50°C) at the studied experimental conditions (C_0: 5 mg/L, *m*: 25 g/L, *t*: 30 min, pH: 5.6), were fitted with the Freundlich and Langmuir isotherm models. The isotherm parameters and their linear correlation coefficients are given in Table 2. The experimental data fit well to the Langmuir and Freundlich isotherm models. However, the Freundlich model gives a slightly better fit than the Langmuir model. This agrees with the report of Mortula and Gagnon [8]. They revealed that the adsorption of phosphate onto the alum sludge was best described by the Freundlich isotherm.

As can be seen from Table 2, when the temperature is increased from 10°C to 40°C, the $q_{\max}$ values increased from 0.696 to 0.760 mg/g. This result suggested that the adsorption of phosphate on the alum sludge is endothermic. Besides, it can be said that, as stated by Kondapalli and Mohanty [26], the increase of sorption capacity with increased temperature is due to the increase of the kinetic energy of the sorbent particles. Hence, as a result of the increased collision frequency between sorbent and sorbate, sorption onto the surface of the sorbent is enhanced. Bond rupture at high temperatures may lead to an increase in the number of active sorption sites. Accordingly, sorption may be enhanced [26].

Table 2
Langmuir vs. Freundlich isotherm parameters

Temperature (°C)	Langmuir isotherm			Freundlich isotherm		
	$q_{\max}$ (mg/g)	*b* (L/mg)	R^2	K_F (mg/g)	1/*n*	R^2
10	0.696	0.075	0.946	0.062	0.630	0.994
20	0.701	0.117	0.987	0.088	0.587	0.990
30	0.754	0.143	0.984	0.114	0.555	0.962
40	0.760	0.232	0.989	0.199	0.384	0.991
50	0.717	0.104	0.976	0.096	0.578	0.972

From Table 2, it can be seen that the values of the Langmuir isotherm constant (*b*) and Freundlich constant (K_F) increase with an increase in temperature, except for 50°C. These increases can be attributed to the endothermic nature of the adsorption process as well [27]. The values of $1/n$ less than 1 demonstrate favorable sorption.

3.5. Comparison of the adsorption capacity with different adsorbents

In this study, the maximum phosphate adsorption capacities of various adsorbents in the literature and the alum sludge used in this experiment were compared and presented in Table 3.

As can be seen from Table 3, the adsorbents such as crawfish char derived at different pyrolysis temperatures, red mud, fly ash and nano-alumina having particle size <50 nm of Sigma-Aldrich had lower phosphate adsorption capacity than that of the alum sludge used in this study. Moreover, the alum sludge exhibited a higher adsorption capacity of phosphate than that of the activated charcoal from Fisher Scientific Ltd. This suggests that the local alum sludge seems to be a promising adsorbent due to the high adsorption capacity compared to some adsorbents, especially commercial activated charcoal. Besides, alum sludge is a by-product that is easily available and free of charge. However, it can be realized from Table 3 that the maximum adsorption capacity of the alum sludge used in this study was found to be lower than those of the local alum sludges from various countries [6,8–10]. This trend, as was previously mentioned, is attributed to the chemical composition of the sludge, which varies depending on the raw water quality and origin, the soil type of the region, nature of the chemicals used in the treatment plant [2,3] and the dosage applied [4]. Furthermore, the difference in the experimental conditions (i.e. adsorbent dosage, initial pH, temperature, contact time), as well as the chemical compositions of the alum sludge may be the reason for the differences of phosphate adsorption capacities.

3.6. Adsorption thermodynamics

The effect of solution temperature was studied in various concentrations of the phosphate at the range of 10°C–40°C. Data is presented in Fig. 6. As can be seen from Fig. 6, when the temperature increased from 10°C to 40°C, the phosphate adsorption capacity also increased. This result is attributed to the endothermic nature of the adsorption process at

Table 3
Comparison of the adsorption capacities of phosphate onto the alum sludge with different adsorbents reported in the literature

Adsorbent	q_{max} (mg/g)	Reference
Alum sludges from various local water treatment plants		
Alum sludge (Lake major water treatment plant in Halifax, Canada)	1.03	[6]
Alum sludge (Mannheim water treatment plant in Ontario, Canada)	1.11	[6]
Alum sludge (Lake major water treatment plant in Halifax, Canada)	7.11	[8]
Alum sludge (Ballymore-Eustace waterworks located in Co. Kildare, South Dublin, Ireland)	10.02	[9]
Alum sludge (18 months aging sludge) (Ballymore-Eustace waterworks located in Co. Kildare, South Dublin, Ireland)	0.90–23.90	[10]
Low-cost adsorbents		
Activated rice husk ash	0.74	[18]
Crawfish char derived at different pyrolysis temperature	0.12–4.21	[20]
Carbonized sewage sludge	0.51–0.82	[28]
Fe–Mn oxide adsorbent	0.99–1.00	[29]
Raw (unmodified) peat	0.92	[25]
Iron-modified peat	11.53	[25]
Ferric sludge	30.00	[30]
Synthetic iron oxide coated sand	1.50	[31]
Naturally iron oxide coated sand	0.88	[31]
Iron oxide coated crushed brick	1.75	[31]
Industrial solid waste Fe(III)/Cr(III) hydroxide	6.53	[32]
Red mud	0.58	[33]
Steel furnace slag	1.43	[34]
Fly ash	0.10–0.20	[35]
Commercial activated carbons		
Nano-alumina having particle size <50 nm of Sigma-Aldrich	0.16	[36]
Activated charcoal from Fisher Scientific Ltd.	0.46	[36]
Alum sludge, collected from a drinking water treatment plant in Istanbul, Turkey	0.67–0.76	This study

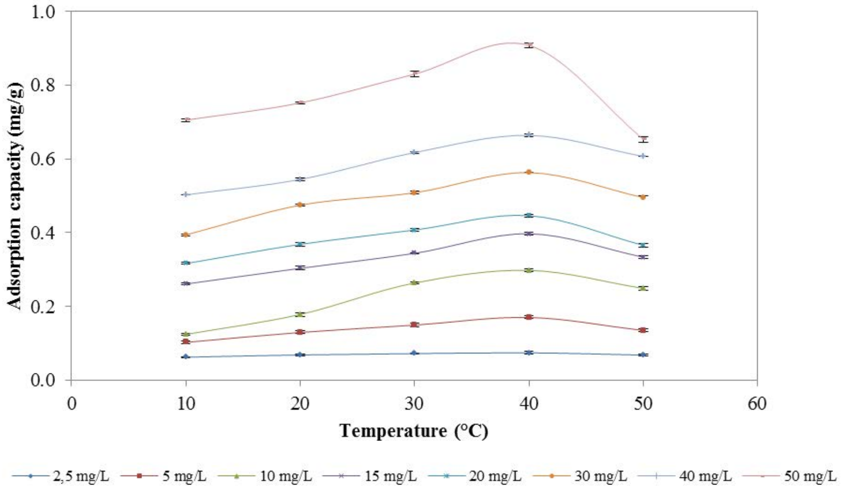

Fig. 6. Effect of temperature on adsorption of phosphate ions on alum sludge (C_0: 5 mg/L, *m*: 25 g/L, *t*: 30 min)

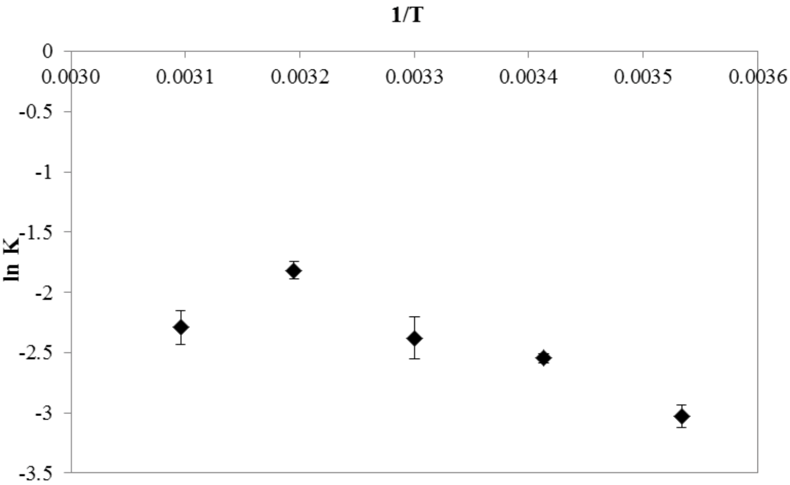

Fig. 7. Relationship plot between lnK_c against 1/*T* of the adsorption of phosphate on alum sludge.

temperatures ranging from 10°C to 40°C. It can be said that, as stated by Chowdhury [27], an increase in temperature leads to an increase in the number of accessible active sites on the adsorbent surface, an increase in the porosity, and increase in the pore volume of the adsorbent. Besides, temperature increase also causes an increase in the rate of diffusion of the adsorbate molecules across the external boundary layer and in the internal pores of the adsorbent particle. Consequently, an increase in adsorption may be obtained. However, as can be seen from Fig. 6, the phosphate adsorption capacity slightly decreases above 40°C.

In Fig. 7, the relationship plot between ln*K* against 1/*T* of the adsorption of phosphate on alum sludge is shown. As known, the plot of ln*K* vs. 1/*T* gives the numerical value of Δ*H* of the adsorption of phosphate; Δ*G* and Δ*S* are calculated.

Thermodynamic parameters for phosphate adsorption on alum sludge are given in Table 4. The positive values of Δ*G* indicate that the adsorption was not spontaneous. The positive value of Δ*H* (17.1 kJ/mol) shows that the adsorption of phosphate on alum sludge is an endothermic reaction. The value of Δ*H* also suggests that phosphate adsorption on alum sludge is a physical adsorption process. As pointed out by Saha and Chowdhury [37], the heat produced during physical adsorption is of the same order of magnitude as the heats of condensation (2.1–20.9 kJ/mol), and a positive value of Δ*S* (36.49 kJ/mol) indicates the affinity of the adsorbent towards the adsorbate species, increased randomness at the

Table 4
Thermodynamic parameters for phosphate adsorption on alum sludge

Temperature (K)	Δ*G* (kJ/mol)	Δ*H* (kJ/mol)	Δ*S* (kJ/mol)
283	7.12	17.10	36.49
293	6.20		
303	5.99		
313	4.73		
323	6.15		

solid/solution interface with some structural changes in the adsorbate and the adsorbent, and an increase in the degree of freedom of the adsorbed species.

It can be seen from Table 4 that, ΔG values in the range of temperatures between 283 and 313 K decrease with an increase in the temperature, which accounts for a decrease in the feasibility of adsorption at higher temperatures [38]. A similar behavior (positive value of ΔG, ΔH, and ΔS) for the effect of temperature on phosphate adsorption was observed for the Fe–Mn oxide adsorbent [29], biochars produced by the pyrolysis of sugar cane bagasse and *Miscanthus × giganteus* [39].

4. Conclusions

The results indicate that the local alum sludge in Istanbul can be used as an adsorbent for phosphate removal from aqueous solution. It was seen that the adsorption capacity, q (mg/g), of the alum sludge was dependent on the initial pH and temperature, and increased at the pH range of 2–6 and the temperature range of 10°C–40°C. The adsorption capacity increased at the adsorbent dosage of 5–30 mg/L. The maximum adsorption capacity, $q_{\max}$ (mg/g), at the temperature range of 10°C–40°C varied from 0.696 to 0.760 mg/g. Data relating to the phosphate adsorption by the alum sludge fitted well to the Freundlich isotherm. Thermodynamic parameters suggested the endothermic nature of the phosphate adsorption and the affinity of the adsorbent for phosphate.

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