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PHOSPHORUS ADSORPTION/DESORPTION KINETICS OF BIORETENTION

by

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A bioretention medium has considerable influence on the removal of pollutants, especially phosphorus pollutants. In this paper, the phosphorus adsorption properties of three bioretention media (sand, iron powder, and aluminum powder) are studied, and the effects of pH and ionic strength on the phosphorus adsorption are analyzed. Results show that the phosphorus adsorption isotherm can be well modeled by the Langmuir equation. The experimental results show the monolayer adsorption capacity of sand is the highest, while the iron powder the lowest. The pH of the solution has a considerable influence on the phosphorus adsorption of sand and iron powder, but has a minimal effect on aluminum powder.

Key words: *phosphorus, adsorption and desorption, pH, ionic strength, geometric potential theory, fractal calculus*

Introduction

As a technical measure used to reduce the urban storm-water and to control the run-off pollution, bioretention has been widely used in urban construction in many countries. Traditional bioretention has a good removal efficiency on particulate phosphorus (P) through precipitation and filtration, but its removal efficiency on the dissolved phosphorus is not so ideal [1]. The phosphorus removal from rainwater through bioretention is accomplished through the sedimentation and absorption of fillers, plants, and microorganisms, with adsorption as the most important method of phosphorus removal [2]. Zhang *et al.* [3] showed that agricultural non-point source pollution causes water quality deterioration, in which the phosphorus output of farmland soil surface runoff leads to river and lake's eutrophication. Hence, the research on the process, mechanism, and control measures of phosphorus loss from farmland soil into water has been receiving increasing attention from scholars. The most commonly used methods for phosphorus removal include crystallization, precipitation, biological phosphorus removal, and adsorption [4]. At present, chemical precipitation and biological methods have better dephosphorization effects and have been frequently applied. Adsorption requires simple equipment, low operating costs, and wide raw material sources [5]. At present, PO_4^{3-} adsorption by different media frequently uses three models, namely, Langmuir, Freundlich, and Temkin models, to simulate the phosphorus adsorption experiment. The selection of adsorption medium has an important influence on adsorption. The commonly used

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porous adsorption media are zeolite, diatomaceous earth, and industrial wastes, such as aluminum iron mud and fly ash [6]. However, research on phosphorus adsorption by using iron and aluminum powders are relatively rare.

In the present study, the properties of phosphorus adsorption and desorption by using sand, iron powder, and aluminum powder are studied. The effects of different initial phosphorus concentrations, solution pH values, and ionic strengths on phosphorus adsorption and desorption by the three media are investigated.

Materials and methods

Experimental materials

The test soil is obtained from the sand near the river in Haojiayao Village, Shuanghe Town, Tuoketuo County, Hohhot in April 2017. The soil in this area is unpolluted, and debris, such as gravel and plant roots, are removed. Then, sand is collected and brought to the laboratory. The sand sample is prepared for later use after 1 mm sieving. The results of sand particle size analysis are expressed as follows: sand (1-0.05 mm) accounts for 90%, powder (0.05-0.01 mm) accounts for 2%, and clay (< 0.01 mm) accounts for 8% according to the international soil texture and these sand types are called sand soil and sandy loam soil. The iron and aluminum powders are purchased from the distribution department of Keyu Experimental Instruments in Xincheng District, Hohhot. Iron and aluminum powders, which are produced by Tianjin Fuchen Chemical Factory and Tianjin Damao Chemical Factory, respectively, are analytically pure ($\geq 99\%$).

Experimental methods

Isothermal adsorption experiment

The phosphorus solutions at different initial concentrations (10, 20, 30, 40, and 50 mg/L) are prepared with monopotassium phosphate (analytical pure: $\geq 99.7\%$), and 100 mL of each solution is taken into 250 mL conical flasks. A total of 2 g sand (iron and aluminum powders) are placed in 250 mL conical flasks and shaken at a certain temperature (25 °C) for 36 hours. Then, the samples are centrifuged at 4000 rpm for 10 minutes. The supernatant is obtained and then filtered through a 0.45 μm filter paper, and the phosphorus concentrations in the solutions are measured. The phosphorus concentration in the solution is determined by ammonium molybdate spectrophotometry.

Desorption experiment

After the isothermal adsorption experiment, six samples, namely, sand, iron powder, and aluminum powder, with initial phosphorus concentrations of 10 mg/L and 30 mg/L, are used, and the samples are replaced with distilled water. Sand, iron powder, and aluminum powder are sampled after shaking at a certain temperature (25 °C) for 12, 24, 48, 60, and 72 hours. Then, the samples are centrifuged at 4000 rpm for 10 minutes. The supernatant is obtained and then filtered through a 0.45 μm filter paper, and the phosphorus concentrations in the solution at different times are measured.

The phosphorus adsorption kinetics experiments

A total of 2 g sand, iron powder, and aluminum powder into are placed in 250 mL conical flasks, and 100 mL of phosphorus solution with the mass concentration of 5 mg/L,

which is prepared by 0.02 mol/L NaCl solution, is added. The samples are shaken at a constant temperature of 25 °C with the speed of 180 rpm. They are sampled every 1 hours, and the samples are centrifuged at 4000 rpm for 10 minutes. The supernatant is obtained and then filtered through a 0.45 μm filter paper, and the phosphorus concentrations in the solutions are measured. According to the changes in phosphorus concentration, the adsorption amount of the media to phosphorus is calculated, and the phosphorus adsorption kinetic characteristics curves of the media are drawn. The adsorption amount is calculated:

$$q_t = \frac{(C_0 - C_t)v}{m} \tag{1}$$

where q_t [mgg^{-1}] is the amount of phosphorus adsorbed by the medium at time t, C_0 [mgL^{-1}] – the initial phosphorus concentration, C_t [mgL^{-1}] – the phosphorus concentration in the supernatant at time t, V [L] – the volume of the solution, and m [g] – the medium mass.

The phosphorus adsorption experiment under different influencing factors

- *The pH.* A total of 2 g sand, iron powder, and aluminum powder are placed into the 250 mL conical flasks, and 100 mL of phosphorus solution with the mass concentration of 30 mg/L is added into the conical flasks. The solutions are adjusted with 0.1 mol/L HCl and 0.1 mol/L NaOH, and the pH of the solutions are 4, 5, 6, 7, 8, 9, 10, and 11. At the temperature of 25 °C, the samples are shaken for 36 hours and centrifuged for 10 minutes. Then, the supernatant is obtained and then filtered through a 0.45 μm filter paper, and the phosphorus concentrations in the solutions are measured.
- *Ionic strength.* A total of 100 mL of phosphorus solution with the mass concentration of 30 mg/L, 2 g sand, 2 g iron powder, and 2 g aluminum powder are added into 250 mL conical flasks. The solutions with different ionic strengths of 1, 5, 10, and 20 mmol/L are prepared with KCl. At the temperature of 25 °C, the samples are shaken for 36 hours, centrifuged for 10 minutes, and filtered through a 0.45 μm filter paper. Lastly, the supernatant is taken to measure the phosphorus concentrations in the solutions.
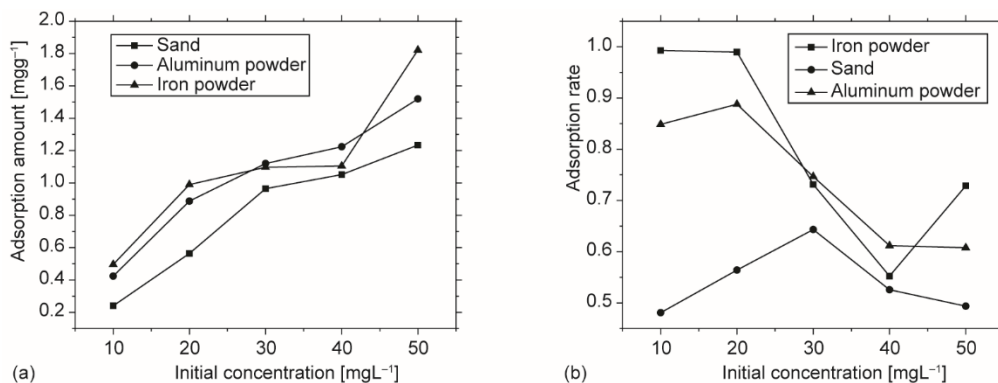
Results and discussion

Isothermal adsorption results of media to phosphorus

The linear expression of the Langmuir isotherm model is expressed:

$$\frac{1}{q_e} = \frac{1}{bq_{max}C_e} + \frac{1}{q_{max}} \tag{2}$$

where q_e [mgg^{-1}] is the unit adsorption amount in the adsorption equilibrium, q_{max} [mgg^{-1}] – the theoretical single-layer saturated adsorption amount, b [mg^{-1}] – the equilibrium constant, and C_e – the equilibrium concentration of the adsorbate.

The results of isothermal adsorption of various media to phosphorus is shown in figs. 1(a) and 1(b). The fitting results of Langmuir equation are shown in tab. 1. As shown in tab. 1, the relationship among the monolayer adsorption capacities of sand, iron powder, and aluminum powder is sand > iron powder > aluminum powder, mainly because the specific surface area of sand is larger than those of iron and aluminum powders. Hence, sand can provide additional adsorption points for phosphorus, thereby increasing the adsorption amount of phosphorus by sand [7]. Second, solid $CaCO_3$ may exist on the soil surface, and the adsorption and immobilization effect on phosphorus is extremely strong, which enable the sand to

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Figure 1. Isothermal phosphorus adsorption curve of each medium; (a) absorption amount of different media, (b) absorption rate of different media

increase the adsorption amount of phosphorus [8]. For metals, iron and aluminum powders have strong phosphorus adsorption, but the adsorption capacity of iron power is higher than that of aluminum powder. The dimensionless adsorption strength, R_L, of the three media is $0 < R_L < 1$, which indicates that phosphorus adsorption is feasible [9].

Table 1. Langmuir isothermal adsorption parameters for phosphorus absorption of different media

Adsorbent	q_{max} [mgg^{-1}]	B [mg^{-1}]	R_L	Equation	R^2
Sand	2.9163	0.0322	$0 < R_L < 1$	$y = 10.64x + 0.3429$	0.7513
iron powder	1.3328	9.4615	$0 < R_L < 1$	$y = 0.0793x + 0.7503$	0.9072
aluminum powder	2.5849	0.0062	$0 < R_L < 1$	$y = 2.316x + 0.543$	0.8398

As shown in fig. 1(a), the adsorption amount of sand, iron powder, and aluminum powder increases with the increase in initial concentration, thereby indicating that the phosphorus concentration of the solution is a main factor affecting adsorption. First, the higher the concentration of the solution is, the higher the available phosphorus is for the medium to adsorb. Second, the higher the concentration of the solution is, the greater the concentration difference between the bulk of the solution and the liquid film on the outer surface of the medium will be, thereby resulting increasing the mobility of phosphorus to the surface of the medium [10]. Therefore, as the phosphorus concentration in the solution increases, the amount of phosphorus adsorbed by the medium increases. As shown in fig. 1(b), the adsorption rate of sand and aluminum powder increases with the increase in initial concentration. When the mass concentrations of sand and aluminum powder solutions reach 30 and 10 mg/L, respectively, the phosphorus adsorption rate of the two media begin to decrease, which is consistent with the findings of Del Bubba *et al.* [11]. The phosphorus adsorption rate by iron powder decreases first slowly with the increase in phosphorus concentration and decreases rapidly after reaching a certain concentration. As the phosphorus concentration continues to increase, the adsorption rate has a tendency to increase again. The optimum initial concentration conditions for sand, iron powder, and aluminum powder removal rates are 30, 10, and 20 mg/L, and their maximum adsorption rates reach 64.32%, 99.27%, and 88.79%, respectively.
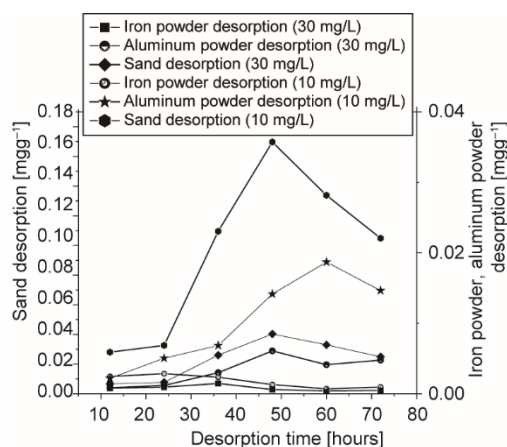
The phosphorus desorption results in different media are shown in fig. 2. The figure shows the changes in the desorption amount of phosphorus by sand, iron powder, and alumi-

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num powder with time. The change trends of
phosphorus desorption by sand and aluminum
powder in the media are almost the same.
Sand and aluminum powder with the initial
concentration of 30 mg/L and sand with the
initial concentration of 10 mg/L have the
same phosphorus desorption trend. Within 12-
48 hours, the desorption amount of phospho-
rus by the media continues to increase with
the continuation of time, thereby reaching a
maximum at 48 hours, and the maximum val-
ues are 0.1698, 0.0289, and 0.0695 mg/g.
Subsequently, the desorption amount of phos-
phorus by sand shows a decreasing trend with
time. However, the phosphorus desorption by
aluminum powder increases again within 60-
72 hours, which shows that the phosphorus
adsorption-desorption equilibrium is a long

**Figure 2. The phosphorus desorption curves in
different media;** *Note: The concentration in the
parentheses after the label represents the initial
concentration*

dynamic process. For the phosphorus solution with the initial concentration of 10 mg/L and in
the time range of 12-60 hours, the desorption amount of phosphorus by aluminum powder in-
creases slowly with time, thereby reaching the maximum at 60 hours, and the maximum value
is 0.0187 mg/g. A reduced state is shown in the time range of 60-72 hours. At the initial con-
centrations of 10 mg/L and 30 mg/L and in the time range of 12-24 hours, the desorption
amount of phosphorus by iron powder increases with time, but it reaches its maximum value
first at low initial concentration, which is 0.0029 mg/g. Meanwhile, the desorption amount of
phosphorus by iron powder at the high initial concentration continues to increase with time,
thereby reaching its maximum value of 0.0070 mg/g at 36 hours. Afterward, both show a de-
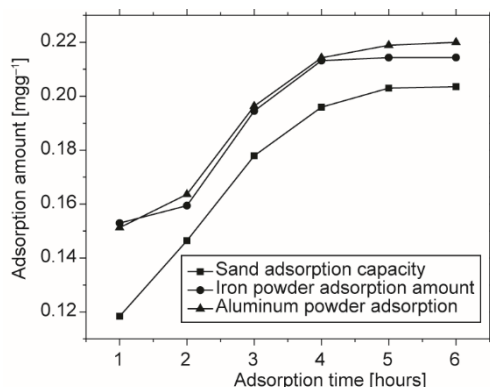creasing tendency with time.

Under the same initial concentration, the relationship among the maximum desorp-
tion amounts of the three media is as follows: sand > aluminum powder > iron powder. The
desorption amount of phosphorus in the three media shows that the desorption amount at high
concentration is higher than that at low concentration, thereby indicating that the initial con-
centration has a certain influence on the desorption amount of phosphorus. The higher the ini-
tial concentration is, the larger the desorption amount of phosphorus in the medium will be.
When the medium has a relatively high phosphorus content, the surface of the solid medium
dissolves to release a part of the physically adsorbed phosphorus as the desorption time con-
tinues, which finally reaches a dynamic equilibrium. This result shows that in the high phos-
phorus environment, the adsorption points of the medium are increased, thereby increasing
the adsorption capacity of the soil [12, 13]. In the low phosphorus environment, the soluble
phosphorus content in the medium is increased, thereby promoting phosphorus desorption in
the medium [12, 14].

Results of kinetic experiments of phosphorus adsorption by media

The phosphorus adsorption kinetics of sand, iron powder, and aluminum powder is
shown in fig. 3. At the initial stage of adsorption, the phosphorus adsorption amount by each
medium increases rapidly with time and then tends to be gentle. The three media in this experi-
ment essentially reach their own adsorption equilibrium after 6 hours. The relationship

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Figure 3. The phosphorus adsorption kinetics curve

among the equilibrium adsorption capacities of the three media is expressed as follows: aluminum powder (0.22 mg/g) > iron powder (0.214 mg/g) > sand (0.204 mg/g). The figure shows that aluminum and iron powders have good adsorption effects on phosphorus.

Generally, the adsorption kinetics of the solute in the solution by the adsorbent can be described by first- and second-order kinetic models.
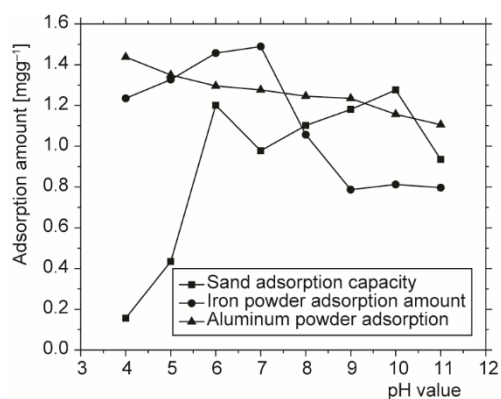
The commonly used expression for first-order kinetics is:

$$q_t = A \left(1 - e^{-k_1 t}\right) \tag{3}$$

The quasi-secondary kinetic model is expressed:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{4}$$

where q_t is the adsorption amount of phosphorus by the medium, t – the adsorption time, k_1 – the first-order kinetic rate constant, A – the constant related to initial mass concentration, k_2 – the quasi-second-order kinetic constant, and q_e – the equilibrium adsorption amount. As shown in eq. (3), for the first-order kinetic model, when time $t \to +\infty$, $q_t \to A$, A approximates q_e and the equilibrium phosphorus adsorption amount by the medium.

As shown in tab. 2, the first- and quasi-second-order kinetic models can reflect the adsorption kinetics of phosphorus by aluminum powder, and the equilibrium adsorption amount is 0.253 mg/g. The phosphorus adsorption by sand and iron powder agrees with the quasi-second-order kinetic model, and the correlation coefficient is > 0.99, thereby indicating that chemical adsorption is the control step during phosphorus adsorption by the three media [15]. The equilibrium phosphorus adsorption amounts by sand and iron powder are 0.247 and 0.245 mg/g, respectively.

Table 2. First-order and quasi-secondary kinetic fitting parameters of phosphorus adsorption by different media

Medium type	First-order kinetic model			Quasi-secondary kinetic model		
	R^2	A	K_1 [Lmg^{-1}h^{-1}]	R^2	q_e [mgg^{-1}]	K_2 [Lmg^{-1}h^{-1}]
Sand	0.8976	0.4334	0.000774	0.9952	0.2470	0.0576
Iron powder	0.8521	1.0624	0.000230	0.9908	0.2451	0.0884
Aluminum powder	0.9206	0.1315	0.003963	0.9936	0.2527	0.0795

Factors affecting phosphorus adsorption

Effect of pH on phosphorus adsorption by media in solution

The variation curves of the phosphorus adsorption capacities of sand, iron powder, and aluminum powder in the solutions with different pH are shown in fig. 4. The change in pH of the solution has minimal effect on the phosphorus adsorption amount of in aluminum
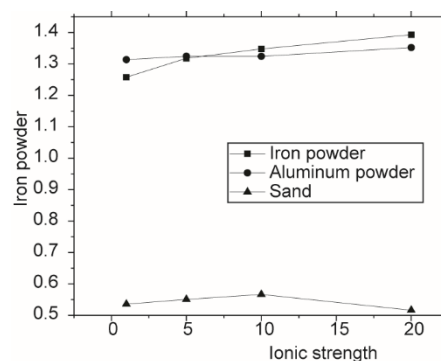
powder, and that of iron powder and sand is substantially affected by the change in pH of the solution. The phosphorus adsorption amount by aluminum powder shows a slow decreasing trend with the increase in pH of the solution, thereby indicating that an acidic environment promotes phosphorus adsorption by aluminum powder. The phosphorus adsorption amount by iron powder increases first with the increase in the pH of the solution. When the pH is 7, the phosphorus adsorption amount by iron powder reaches the maximum value of 1.489 mg/g. With the increase in pH of the solution, the phosphorus adsorption amount by iron powder shows a decreasing trend. This result indicates that a neutral environment is the most beneficial for

Figure 4. Effect of pH on phosphorus adsorption;
Note: The concentration in the parentheses after the label represents the initial concentration

iron powder to adsorb phosphorus. For sand, the pH of the solution has a considerable influence on phosphorus adsorption. When the pH of the solution is 4 to 6, the amount of phosphorus adsorbed by sand increases rapidly with the increase of pH. When the pH of the solution ranges from 6 to 10, the amount of phosphorus adsorbed by sand tends to be stable. Subsequently, the amount of decreases first with the increase in the pH of the solution. It indicates that a neutral or an alkaline environment contributes to the adsorption of phosphorus by sand. This result indicates that a neutral or an alkaline environment contributes to phosphorus adsorption by sand because for the phosphoric acid of tribasic acid that is easily hydrolyzed, different forms of phosphorus, such as $H_2PO_4^-$, HPO_4^{2-}, and PO_4^{3-}, exist in the water body, and phosphorus exists in different forms under different pH conditions, thereby affecting phosphorus adsorption by the media. When pH < 7.2, $H_2PO_4^-$ is the main form of phosphorus. When $7.2 <$ pH < 12.2, HPO_4^{2-} is dominant. At pH = 7.20, phosphorus exists mainly in form $H_2PO_4^-$ and HPO_4^{2-} coexistence. Under the conditions of weak acid or neutral environment, namely, pH ranging from 4 to 7, $H_2PO_4^-$ is the main form of phosphorus [10]. This theory can be used to explain the results of our experiments, thereby indicating that phosphorus adsorption by sand and iron powder is mainly in the form of HPO_4^{2-}, and phosphorus adsorption by aluminum powder is mainly $H_2PO_4^-$. However, when the pH increases, the OH^- concentration increases, OH^- can replace PO_4^{3-} anion, and PO_4^{3-} and OH^- compete for adsorption [16], thereby decreasing the phosphorus adsorption amount by the three media.

Effect of ionic strength

The results of phosphorus adsorption by sand, iron powder, and aluminum powder under different ionic strengths are shown in fig. 5. The adsorption amount of phosphorus by the three media slightly changes under varying ionic

Figure 5. Effect of ionic strength on phosphorus adsorption

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strength. The adsorption amount of phosphorus by iron and aluminum powders shows a continuously increasing trend with the increase in ionic strength, thereby indicating that the larger the ionic strength is, the better the phosphorus adsorption by iron and aluminum powders will be. When the ionic strength increases from 0 mmol/L to 20 mmol/L, the adsorption capacities of iron and aluminum powders increase by 0.135 and 0.038 mg/g, respectively. The increase in the adsorption capacity of iron powder is 3.55-fold higher than that of aluminum powder, thereby indicating that iron powder is more affected by ionic strength in terms of phosphorus adsorption. When an electrostatic attraction is present between the adsorbent and adsorbate, increasing the ionic strength has detrimental effects for adsorption. When electrostatic repulsion is observed between the adsorbent and adsorbate, increasing the ionic strength is beneficial for adsorption [17]. This result indicates electrostatic repulsion between iron powder and PO_4^{3-} and between aluminum powder and PO_4^{3-}. When the ionic strength is in the range of 0-10 mmol/L, the phosphorus adsorption amount by sand increases with the increase in ionic strength. When the ionic strength is 10-20 mmol/L, the amount of phosphorus adsorbed by sand decreases with the increase in ionic strength. When the ionic strength is 10 mmol/L, the amount of phosphorus adsorbed by sand reaches the maximum value of 0.567 mg/g. This result indicates that electrostatic repulsion is also present between sand and PO_4^{3-}. However, although the medium sand does not form an ion pair, the interaction between ions in the medium sand and other ions increases with the increase in ionic strength, and the effective concentration of ions in the sand decreases, which also decreases the amount of phosphorus adsorption [18-20].

Conclusions

This paper studied phosphorus adsorption kinetics experimentally, however its mechanism can be explained by the geometric potential theory [21-24], and a fractal modifications for Langmuir, Freundlich, and Temkin models can be further studied by the fractal calculus [25-28], for example, Liu, *et al.* [28] suggested a fractal rate model for adsorption kinetics, and Fan *et al.* [29] suggested a fractal model for the bio-mechanical properties of wools with great success. This paper gives the following conclusions:

- The quasi-secondary kinetic model is more suitable for the phosphorus adsorption kinetics model of sand, iron powder, and aluminum powder than the first-order kinetic model, thereby indicating that chemisorption is the control step in the phosphorus adsorption by the three media. The isotherm phosphorus adsorption curves of the three media are consistent with Langmuir equation. The fitting results show that the relationship among the maximum adsorption capacities of the three media is expressed as follows: sand > aluminum powder > iron powder.

- The isothermal adsorption experiment shows that the phosphorus adsorption amount by sand, iron powder, and aluminum powder continuously increases with the increase in the initial concentration. However, in terms of phosphorus removal efficiency, when the phosphorus concentration in the solution reaches a certain level, the phosphorus removal efficiency of the medium begins to decrease. The maximum removal efficiencies of sand, iron powder, and aluminum powder are 64.32%, 99.27%, and 88.79%, respectively.

- The desorption amounts of sand, iron powder, and aluminum powder are constantly changing. The higher the initial phosphorus concentration in the solution is, the larger the desorption amount of the medium will be, which indicates that the adsorption points of the medium are increased under high phosphorus environment. This result promotes the adsorption capacity of the medium. Under low phosphorus environment, the soluble

phosphorus content in the medium is increased, which promotes phosphorus desorption by the medium.
- The phosphorus adsorption performance of sand and iron powder is considerably affected by the change in pH, while that of aluminum powder is minimally affected. The best pH for phosphorus adsorption by sand, iron powder, and aluminum powder are 6, 7, and 4, respectively.
- The changes in ionic strength have minimal effects on the adsorption performances of sand, iron powder, and aluminum powder. The optimum ionic strengths for sand, iron powder, and aluminum powder are 10, 20, and 20 mmol/L, respectively. Iron powder is more affected by ionic strength in terms of phosphorus adsorption than aluminum powder.

Acknowledgment

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