ISOTHERMAL ADSORPTION EXPERIMENT OF NITRATE NITROGEN IN BIORETENTION MEDIA

by

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Bioretentions can remarkably improve the rainwater quality. Though iron and aluminum as fillers in bioretentions can remove nitrate nitrogen, its adsorption mechanism is not clear yet and was rarely studied. This paper takes sand, iron powder, and aluminum powder as the research objects to study the adsorption characteristics of nitrate nitrogen in the bioretention with different pH values and different ionic strength. The results show that aluminum powder has the highest the adsorption capacity. Langmuir adsorption isotherm equation and the quasi-secondary kinetic model are adopted to analyze the experimental data.

Keywords: *bioretention, nitrate nitrogen, adsorption, pH, ionic strength*

Introduction

In recent years, the rapid development of cities has led to the replacement of impervious ground and buildings with low-lying and permeable soils [1]. Rainwater runoff is an important way for pollutants to move from impervious grounds to surface water [2]. The removal of pollutants from rainwater runoff by bioretentions is a relatively new but promising technology [3]. Because it can improve the rainwater quality [4], it has a good effect on removing total suspended solids (TSS), heavy metals, phosphorus and bacteria in rainwater runoff [2, 4, 5].

Bioretentions generally include cover, plant, soil, gravel, and drainage layers. Among them, the soil layer plays an important role in removing pollutants [1], so it is very important to choose a suitable soil layer medium. Generally, the traditional bioretention chooses sand and soil as a filler medium, but the common systems now also choose media with strong adsorption capacity and good permeability, such as wood chips, bark, zeolite, vermiculite, *etc.* [6]. Studies have shown that the removal effect of nitrate nitrogen and organic nitrate nitrogen in bioretentions is relatively good, but its removal process is unstable due to the appearance of nitrate nitrogen leaching [5, 7]. In the conventional bioretention, the removal rate of nitrate nitrogen can only reach 29% [8]. Because in the bioretention, the removal of nitrate nitrogen is mainly based on denitrification. The biological denitrification process requires consistent detection and provision of organic substrates. Compared with chemical pro-

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cesses, biological processes are slow and partially incomplete. Adsorption method is recognized as one of the most effective methods to remove nitrate nitrogen [9], and the nitrate nitrogen of some groundwater or surface water is the main problem affecting human health and environment [10]. Therefore, it is necessary to study the adsorption performance of soil layer media to nitrate nitrogen and its influencing factors.

This paper evaluates the adsorption effect of several bioretention media to optimize a bioretention medium for practical applications. In this study, Langmuir isothermal model and Freundlich isothermal model are used to determine the adsorption capacity of several typical media (sand, aluminum powder, and iron powder). In addition, pH values and ion concentrations are also considered as influencing factors for adsorption, and the adsorption kinetics is adopted for the analysis.

Materials and methods

The soil layer media used in this experiment include iron powder, aluminum powder and sand. The sand was taken from the less disturbed area around Hohhot City, China, and the sand was analyzed. The soil used in the experiment was sandy loam. The soil layer media of the bioretention is described in the tabs. 1 and 2.

Table 1. Test materials

Reagent name	Molecular formula	Purity	Manufacturer
Iron powder	Fe	Analytically pure	Changsha Shengyang Chemical Materials Co., Ltd.
Aluminum powder	Al	Analytically pure	Shanghai Deyude Trading Co., Ltd.

Table 2. Data analysis of sand after treatment

Particle size [mm]	1-0.05	0.05-0.01	< 0.001	Texture name
Content	90%	2%	3%	Sandy loam

Adsorption and desorption experiments of nitrate nitrogen

Isothermal adsorption experiment

Sodium nitrate was used to prepare sodium nitrate solutions with different initial concentrations (0, 10, 20, 30, 40, and 50 mg/L), an Erlenmeyer flask was used in the experiment and an iron powder and an aluminum powder were used which were put in a conical flask for shaking 24 hours and centrifuging at 4000 rpm for 10 minutes, and filtering particles larger than 0.45 μm. The concentration of nitrate nitrogen in the solution was determined by phenol disulfonic acid spectrophotometry.

Adsorption of nitrate nitrogen by bioretention media under different pH conditions

Three samples were considered, *e.g.*, sand, iron filings, and aluminum powder, in our experiment, each sample was 2 g in weight, and an optimal concentration was prepared for the adsorption of nitrate nitrogen. The solutions are adjusted with 0.1 mol/L HCl and 0.1 mol/L NaOH, and the pH of the solutions are 3, 4, 5, 6, 7, 8, and 9. The prepared solutions were then shaken for 24 hours at a constant temperature of 25 ° C in a shaking box, and then centrifuged for 10 minutes. The concentration of nitrate nitrogen in the solution was determined by phenol disulfonic acid spectrophotometry.

Adsorption of nitrate nitrogen by bioretention media under different ionic strength conditions

Take three 250 mL conical flasks and add 2 g sand, iron filings, and aluminum powder, respectively, and add the initial concentration that has the best adsorption concentration for nitrate nitrogen. Then, different NaCl solutions with concentrations of 1, 5, 20, 50, and 100 mmol/L, respectively, were added into different media, which were then set in a shaking box, respectively, at 25 °C for 24 hours to reach adsorption equilibrium, and centrifuged for 10 minutes. Finally a filter paper with a pore size of 0.45 μm was used to filter the resultant solutions. The concentration of nitrate nitrogen in the solution was determined by phenol disulfonic acid spectrophotometry.

Kinetic adsorption experiment of nitrate nitrogen in bioretention

Take three 100 mL conical flasks to place 2 g sand, iron filings, aluminum powder, respectively, add 100 mL of sodium nitrate solution with a prepared nitrate nitrogen concentration of 2 mg/L (calculated as N). Shake at 25 °C, 180 rpm, and take samples every hour. After centrifugation for 10 minutes, the supernatant obtained was filtered through filter paper with a pore size of 0.45 μm. According to the change of nitrate nitrogen concentration, the amount of nitrate nitrogen adsorbed by the media is calculated, and the kinetic adsorption curve of the media for nitrate nitrogen is drawn. Calculate the amount of adsorption:

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

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

Desorption experiment of nitrate nitrogen in bioretention

After the isothermal adsorption experiment, weigh 2 g sand, iron powder, aluminum powder and 10 mL plastic centrifuge tube, add 100 mL of distilled water, and sample at 25 °C, 180 rpm speed for 12, 24, 36, 48, and 60 hours. The sample was centrifuged at 4000 rpm for 10 minutes to take the supernatant liquid and filtered through a filter with a pore size of 0.45. Measure the concentration of nitrate nitrogen in the solution to calculate the desorption amount of nitrate nitrogen. In this experiment, only the initial concentration with the best adsorption concentration of nitrate nitrogen was selected for desorption operation. The calculation formula of desorption amount is:

$$Q = \frac{C50}{0.001m} \tag{2}$$

$$k(\%) = \frac{C50}{q_t 100} \tag{3}$$

where Q [mgkg^{-1}] is the desorption amount, C [mgL^{-1}] – the mass concentration of nitrate nitrogen in the filtrate, m [g] – the mass of the substrate, q_t [mgkg^{-1}] – the nitrate nitrogen adsorption capacity of the substrate at time t.

Analysis of experimental results

Isothermal adsorption results of nitrate nitrogen in bioretention

– Langmuir isotherm adsorption model

The linear equation of Langmuir isotherm adsorption is shown in eq. (4):

$$q_e = q_{max}\frac{KC_e}{1+KC_e} \tag{4}$$

where C_e [mgL^{-1}] is the concentration of nitrate nitrogen in the solution when each media is in equilibrium, q_e [mgg^{-1}] – the adsorption amount of each media when it is in equilibrium with nitrate nitrogen, q_{max} [mgg^{-1}] – the maximum adsorption capacity, and K – the constant.

The results of isothermal adsorption of nitrate nitrogen by various media are shown in figs. 1 and 2. The fitting results of Langmuir equation are shown in tab. 3. It can be seen from tab. 3 that the adsorption equilibrium constant, k, value of the three media is sand (0.2212), iron powder (0.0244), and aluminum powder (0.0529). A larger value of k results in a stronger adsorption capacity [11, 12]. Therefore, the relationship between the single-layer adsorption capacity of the three media is aluminum powder > iron powder > sand.

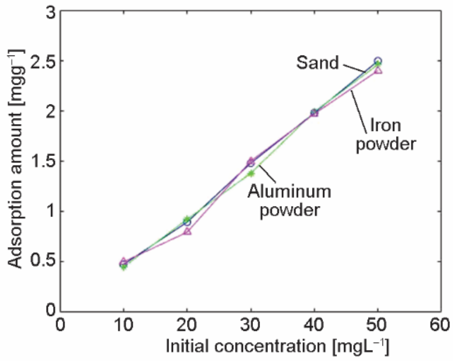

Figure 1. Isothermal adsorption capacity of different media

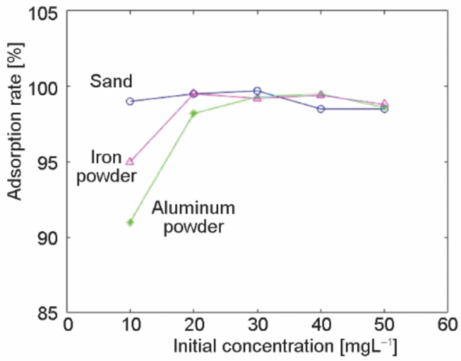

Figure 2. Isothermal adsorption rate of different media

Table 3. Langmuir isotherm adsorption fitting equations and related parameters for adsorption of nitrate nitrate nitrogen in various media

Media	Equation	R^2	q_{max} [mgg^{-1}]	k
Sand	$y = 0.9765x + 0.0207$	0.9970	48.3092	0.0212
Iron powder	$y = 0.9199x + 0.0487$	0.9686	40.4858	0.0244
Aluminum powder	$y = 1.0143x – 0.0247$	0.9997	20.5339	0.0529

As shown in fig. 1, as the initial concentration increases, the adsorption of sand, iron powder, and aluminum powder increases, indicating that the concentration of nitrate nitrogen in the solution is the main factor affecting the adsorption. First, the higher the concentration of the solution, the more the amount of nitrate nitrogen adsorbed by the media. Secondly, as the initial solution of nitrate increases, the driving force of the concentration gradient increases, which results in an increase in the amount of nitrate adsorbed by the media [11]. Therefore, as the concentration of nitrate nitrogen in the solution increases, the amount of nitrate nitrogen adsorbed by the media increases. As shown in fig. 2, as the initial concentration increases, the adsorption rate of sand, iron powder, and aluminum powder increases. When the mass concentration of sand, iron powder and aluminum powder reached 30, 30, and 40 mg/L, respectively, the adsorption rate of nitrate nitrogen by the three media began to decrease. The optimal initial concentration conditions for sand, iron powder, and aluminum powder are 30, 40, and 40 mg/L, and the maximum adsorption rate is 99.7%, 99.5%, and 99.5%.

– Freundlich isotherm adsorption model.

The Freundlich isotherm adsorption can be expressed:

$$q_e = K_f (C_e)^{1/n} \tag{5}$$

where K_f and n are constants, C_e [mgL^{-1}] – the concentration of nitrate nitrogen in the solution at the equilibrium of adsorption of each media, and q_e [mgL^{-1}] – the adsorption amount of each media at the equilibrium of the adsorption of nitrate nitrogen.

Table 2 gives the results of the adsorption of nitrate nitrogen by the three media, where K_f is an adsorbent coefficient [11], a smaller K_f leads to a less adsorption ability, n represents the support of the adsorption process, the greater the n, the stronger the affinity of the adsorbent for the adsorbate in the adsorption process. The more conducive to adsorption [11, 12]. According to the K_f and n values of sand, iron powder, and aluminum powder, the adsorption capacity of the three for nitrate nitrogen is aluminum powder > iron powder > sand. This is consistent with the Langmuir isotherm adsorption equation results. It can be seen from tabs. 1 and 2 that the R^2 of the Langmuir isotherm adsorption equation is greater than the R^2 of the Freundlich isotherm adsorption equation, indicating that the adsorption of nitrate nitrogen by the three media is mainly single-layer adsorption [13], and the three media. The adsorption process of nitrate nitrogen is more in line with the Langmuir isotherm adsorption equation.

Table 4. Freundlich isothermal adsorption fitting equations and related parameters for adsorption of nitrate nitrogen in various media

Media	Equation	R^2	n	K_f
Sand	$y = 1.0701x + 1.027$	0.9968	1.0494	0.9988
Iron powder	$y = 1.4043x + 1.6307$	0.9757	1.0419	1.0169
Aluminum powder	$y = 0.4176x + 0.8797$	0.9987	1.0651	1.0228

Study on the adsorption characteristics of nitrate nitrogen by various media under different pH conditions

The nitrate nitrogen adsorption results of three media with pH values in the range of 3~9 are shown in fig 3. As shown in fig. 3, in the range of pH 3~8, the adsorption capacity of sand increases with the increase of pH. When the pH value is 8~9, the adsorption capacity of sandy soil decreases with the increase of pH. At pH = 8, the adsorption capacity of sand rea-

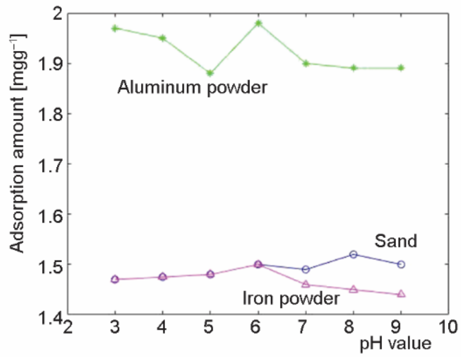

Figure 3. The adsorption curves of various media at different pH values

ches the maximum, which is 1.52 mg/g. It is well-known that the surface charge of colloidal particles is greatly affected the pH value [14]. When the pH value is low, the positive charge distribution in the sand particles is more, and the nitrate nitrogen is negatively charged, which is beneficial to the adsorption of nitrate nitrogen by the sand. With the pH value Increased, the surface of the sand is more negatively charged, and it has an electrostatic repulsion effect with nitrate nitrogen, which reduces the adsorption of sand to nitrate nitrogen. This is consistent with the findings of Xiang *et al.* [14]. In the range of pH value 3~6, the amount of iron powder adsorbing nitrate nitrogen increases. On the one hand, it may be because under acidic conditions, the oxide film on the surface of iron powder is removed, increasing the contact area of nitrate nitrogen and iron powder [15]. On the other hand, speeds up the diffusion process of iron powder into the acidic solution and accelerates the chemical reaction [16]. So that the adsorption of iron powder to nitrate nitrogen increases. when the pH value is 6~9, the adsorption of iron powder shows a decreasing trend. The adsorption capacity of nitrate nitrogen reaches the maximum when pH = 6, which is 1.5 mg/g. Studies have shown that under alkaline conditions, the reaction of iron powder to reduce NO_3^- is very slow, even does not react [15]. When the pH value of aluminum powder is 3~5, the adsorption ability declines greatly due to the possible formation of Al $(OH)_3$ [9], which reduces the adsorption of nitrate nitrogen. When the pH value is 5~6, the adsorption capacity of aluminum powder to nitrate nitrogen increases rapidly. With the increase of pH, the positive charge on the surface of aluminum powder is more, attracting NO_3^-, which leads to an increase in adsorption capacity. When the pH value is 6~9, the aluminum powder adsorbs nitrate nitrogen The adsorption capacity decreases with the increase of pH, which may be due to the competitive adsorption between OH^- and NO_3^-, as well as the increased negative charge on the surface of the aluminum powder, and the electrostatic repulsion on the surface of nitrate nitrogen resulting in a decrease in the adsorption capacity [9]. The adsorption capacity of sand, iron powder and aluminum powder reached the best when the pH was 8, 6, and 6, respectively, and the adsorption capacity was 1.52, 1.97, and 1.5 mg/g.

Study on the adsorption characteristics of nitrate nitrogen in various media under different ionic strengths

The nitrate nitrogen adsorption capacity of the three media under different ionic strengths (NaCl, 1-100 mmol/L) is shown in fig. 4. When the ionic strength is 1-5 mmol/L, the adsorption of nitrate nitrogen by aluminum powder and iron filings decreases with the increase of concentration. When the ion concentration is in the range of 5-100 mmol/L, the adsorption of nitrate nitrogen by aluminum powder and iron filings increases with the increase of concentration. When the ion concentration is 100 mmol/L for aluminum powder and iron filings, respectively, the adsorption capacity of nitrate nitrogen reaches the maximum, which is 1.99 mg/g and 1.54 mg/g, respectively. It is well-known that there is an electrostatic force to enhance the ionic strength, which either decreases or enhances the adsorbtion ability [17]. It shows that there is electrostatic repulsion between aluminum powder and nitrate nitrogen and between iron powder and nitrate nitrogen. When the ion concentration is 1-20 mmol/L, as the ion concentration increases, the adsorption capacity of sand to nitrate nitrogen is reduced. When the ion concentration is in the range of 20-100 mmol/L, sand to nitrate the amount of adsorbed nitrate nitrogen increases with increasing ion concentration. When the ion concentration is 50 mmol/L, the adsorption capacity of sand to nitrate nitrogen reaches the maximum, which is 1.44 mg/g. It shows that there is also electrostatic repulsion between sand and nitrate nitrogen. Although ion pairs cannot be formed in the sand, as the ion concentration increases, the interaction between the ions in the sand and other ions increases, reducing the effective ions in the sand [18], and the amount of nitrate adsorbed decrease. Under different ion concentrations, the maximum changes of the adsorption capacity of nitrate nitrogen by sand, aluminum powder and iron powder are 0.11 mg/g, 0.04 mg/g, and 0.06 mg/g, respectively. It shows that the adsorption capacity of nitrate nitrogen by sand is more affected by the ion concentration.

Desorption experiment results of nitrate nitrogen in bioretention

The relationship between the desorption amount of nitrate nitrogen in three media with time is shown in fig. 5. It can be seen from the figure that the desorption amount of nitrate nitrogen in each media is in the time range of 12-24 hours. As time increases, the desorption amount also increases, between 24 hours and 60 hours, the desorption amount of nitrate nitrogen in each media varies with the increase of time, the balance is basically maintained, and the range of change is gentle. It shows that with the extension of the analysis time, the surfaces of the three media dissolve, releasing part of the nitrate nitrogen physically adsorbed [18], and finally reach equilibrium.

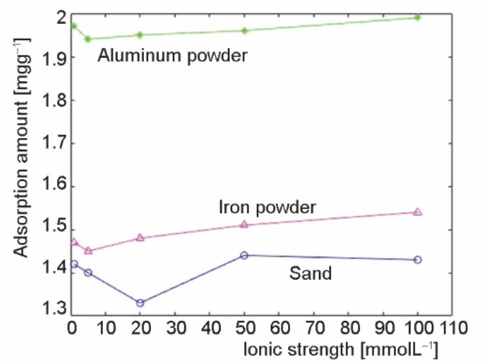

Figure 4. Adsorption curves of various media under different ionic strengths

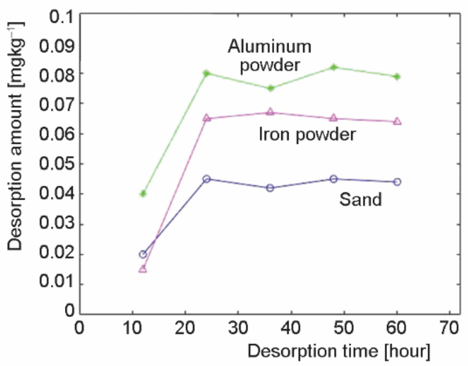

Figure 5. The relationship between the desorption amount of nitrate nitrogen in each media and time

Kinetic adsorption results of nitrate nitrogen in the media of bioretention

The quasi-first-order kinetic model

The quasi-first order kinetic equation can be expressed:

$$q_e - q_t = q_e \exp(-k_1 t) \tag{6}$$

where q_e [mgg^{-1}] is the adsorption capacity of each media when nitrate nitrogen is absorbed, q_t [mgg^{-1}] – the adsorption amount of the media to nitrate nitrogen at time t, t [min] – the reaction of the media adsorption of nitrate nitrogen time, and k_1 [min^{-1}] – the quasi-first order kinetic rate constant.

The quasi-secondary kinetic model

The quasi-secondary kinetic equation is:

$$\frac{1}{q_e - q} = \frac{1}{q_e} + tk_2 \tag{7}$$

where q_e [mgg^{-1}] is the adsorption amount of each media when nitrate nitrogen is absorbed in equilibrium, q_t [mgg^{-1}] – the adsorption amount of the media to nitrate nitrogen at time t, and k_2 – the speed constant of the quasi-secondary kinetic model.

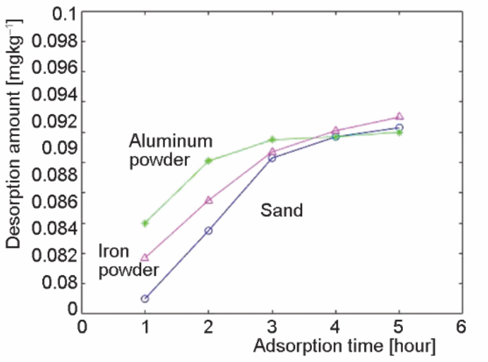

Figure 6. Kinetic graphs of adsorption of nitrate nitrogen by various media

As shown in fig. 6, as the adsorption time increases, the adsorption of nitrate nitrogen by the three media also increases. The entire adsorption process can be divided into two stages of rapid growth and slow growth. In the early stage of adsorption, the growth rate is faster, mainly because there are a large number of active sites on the surface of the media, and the concentration of nitrate nitrogen is high, which is conducive to the mass transfer. As the adsorption progresses, the adsorption sites on the surface of the three media with rapid consumption, the adsorption amount of nitrate nitrogen also becomes slow with the increase of time [12]. The adsorption capacity shows the order of aluminum powder > iron filingsr > sand soil. The first-order kinetic equation and the quasi-second-order kinetic equation are given in tab. 6.

Table 6. First-order kinetic adsorption fitting equations and related parameters for adsorption of nitrate nitrate nitrogen in various media

Media	Equation	R^2	q_e [mgg^{-1}]	k_1
Sand	$y = -0.1656x + 0.2128$	0.9512	1.237	0.1656
Iron powder	$y = -0.2665x + 0.0281$	0.9877	1.028	0.2665
Aluminum powder	$y = -0.1896x + 0.1314$	0.9486	1.140	0.1896

The results are shown in tabs. 6 and 7. In the first-order kinetic model, the R^2 of the three media are sand (0.9512), iron powder (0.9877), and aluminum powder (0.9486). Quasi-second-order kinetics R^2 in the model is sand (0.9997), iron powder (0.9992), and aluminum powder (0.9998). From the correlation coefficients, it can be known that the adsorption process of nitrate nitrogen by these three media is more suitable for fitting with the quasi-second-order kinetic model, which further shows that the adsorption process of nitrate nitrogen by the three media is mainly chemical adsorption.

Table 7. Second-order kinetic adsorption fitting equations and related parameters for adsorption of nitrate nitrogen in various media

Media	R^2	q_e [mgg^{-1}]	K_2	Equation
Sand	0.9997	0.16	26.75	$y = 6.154x + 1.4602$
Iron powder	0.9992	0.16	26.75	$y = 6.1648x + 1.2002$
Aluminum powder	0.9998	0.15	85.45	$y = 6.3853x + 0.5201$

Conclusions

In this paper, the kinetics of nitrate nitrogen adsorption was studied. The results are as follows.

- The quasi-second-order kinetic model is more suitable than the first-order kinetic model with the nitrate nitrogen adsorption kinetic model of sand, iron powder and aluminum powder. Therefore, the adsorption of nitrate nitrogen by the three media is mainly chemical adsorption. The adsorption isotherms of nitrate nitrogen by the three media are consistent with the Langmuir isotherm equation. The results show that the adsorption capacity of the three media for ammonia nitrate nitrogen from high to low is aluminum powder, iron powder, sand.
- The result of isothermal adsorption is that the increase of initial concentration increases the adsorption of nitrate nitrogen by sand, iron powder, and aluminum powder. However, when the concentration of nitrogen nitrate in the solution is at a certain level, the adsorption efficiency begins to decrease. The maximum removal efficiency of sand powder, iron powder, and aluminum powder was 98.5%, 95%, and 91%, respectively.
- The analysis amount of nitrate nitrogen by sand, iron powder and aluminum powder increases rapidly in 12~24 hours. In the range of 24~60 hours, the analysis amount of nitrate nitrogen by the three media is basically saturated. .
- The adsorption performance of sand and iron powder for nitrate nitrogen is less affected by pH changes, while the adsorption performance of aluminum powder is larger. The optimal pH for adsorption of nitrate nitrogen by sand, iron powder and aluminum powder is 8, 6, and 6, respectively.
- The optimal ion concentration of sand, iron powder, and aluminum powder for nitrate nitrogen adsorption is 50, 50, and 1 mmol/L, respectively.

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