ADSORPTION CHARACTERISTICS OF AMMONIUM NITROGEN IN A BIORETENTION SYSTEM

by

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Ammonia nitrogen is one of the important pollutants in urban rainfall. Though iron ions and aluminum ions as a filler for plant retention system have a good effect on the removal of ammonia nitrogen, their adsorption characteristics was rarely studied. This paper uses different media (sand, iron powder, aluminum powder) with different pH values and ionic strengths in the bioretention system to study the adsorption mechanism of ammonia nitrogen in an artificial rainwater. The result shows that Langmuir isothermal adsorption model is more in line with the ammonia nitrogen adsorption process, and the sand has the best adsorption capacity among three samples. Effect of pH values on adsorption kinetics is also studied.

Key words: *bioretention system, ammonia nitrogen, adsorption, pH, ion concentration*

Introduction

Bioretention systems are a promising rainwater management to control water quality [1], and wildly used to treat rainwater on permeable and impervious surfaces in urban areas [2]. Plants, soils and microbial system are used in a bioretention system, which can be applied to various terrains and soil conditions for storage and purification of runoff rainwater by removing phosphorus, suspended solids, chemical oxygen demand and heavy metals [3, 4], and it is not affected by temperature.

The bioretentions system, also known as landscape depressions, is composed of aquifers, vegetation, animal aquifers, overflow structures and optional drainage systems [5]. Absorption and denitrification of plants can remove nitrogen from rainwater before it is discharged to the ground or surface water [6]. In the course of human activities, ammonia nitrogen pollution is serious, which leads to reduce of dissolved oxygen, deterioration of water quality, excessive reproduction of algae and microorganisms, and even eutrophication of water bodies [7]. According to research, the removal rate of ammonia nitrogen by traditional bioretention systems is as high as 74% [8], which is due to the digestion and adsorption process of ammonia nitrogen in the medium layer. Adsorption is a reliable and effective removal

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method. Due to its low energy consumption and simple operation, it is widely used in engineering [9]. This paper studies the adsorption characteristics of soil layer medium to ammonia nitrogen and its influencing factors.

Materials and methods

Experimental materials

In this experiment, experimental soil was obtained on the bank of the Yellow River at the Shenquan Ecological Tourism Base in Hohhot, China. The sand in this area has not been contaminated, and the debris and residual roots are removed, and the samples are naturally dried and brought back to the laboratory. Sand will be ready for use after being screened by 1 mm. According to the international sand texture, gravel (1-0.05 mm) accounts for 90%, powder (0.05-0.01 mm) accounts for 2%, and clay (< 0.01 mm) accounts for 8%. Iron powder and aluminum powder were purchased from Keyu Experimental Instrument Distribution Office, Xincheng District, Hohhot. The iron powder and aluminum powder with analytically pure (≥ 99%) were produced by Changsha Shengyang Chemical Materials Co., Ltd. and Shanghai Deyude Trading Co., Ltd.

Experimental method

Adsorption isotherm experiment

Ammonium chloride was used to prepare ammonium chloride solutions with different initial concentrations (0, 10, 20, 30, 40, 50, 60, 70 mg/L). Each solution of 100 mL was placed in a 250 mL Erlenmeyer flask. Put 2 g of sand (iron powder and aluminum powder) into a conical flask and shake it in a constant temperature (25 °C) shaking box for 24 hours. The sample was then centrifuged at 3000 rpm for 10 minutes. The resulting supernatant was filtered through filter paper with a pore size of 0.45 μm. The concentration of nitrogen in the solution was determined by Nessler's reagent spectrophotometry.

Effect of pH values on adsorption

Put 2 g of, respectively, sand, iron powder, and aluminum powder into a 250 ml Erlenmeyer flask, and add the initial concentration that has the best ammonia nitrogen adsorption concentration. Adjust the pH to 3~9 with 0.1 mol/L HCl and 0.1 mol/L NaOH, respectively. After continuous shaking in a constant temperature shaking box at 25 °C for 24 hours, the filter was allowed to stand, centrifuged for 10 minutes, and the supernatant was filtered through filter paper with a pore size of 0.45 μm. The concentration of ammonia nitrogen in the solution was determined by Nessler's reagent spectrophotometry.

Effect of ionic strengths on adsorption

Weigh 2 g medium (sand, iron powder, aluminum powder) into a 250 mL Erlenmeyer flask and add the initial concentration with the best ammonia nitrogen adsorption concentration. Add NaCl prepared ion concentration 1, 5, 20, 50, and 100 mmol/L solution, respectively. Shake at 25 °C for 24 hours to reach adsorption equilibrium, centrifuge it for 10 minutes, take the supernatant and filter through filter paper with pore size of 0.45 μm. The concentration of ammonia nitrogen in the solution was determined by Nessler's reagent spectrophotometry.

Kinetic adsorption experiment

Weigh 2 g sand, iron shavings, aluminum powder into a 100 mL Erlenmeyer flask, add the initial concentration of ammonia nitrogen adsorption concentration is the best. Shake at a constant temperature of 25 °C, take samples each hour, and centrifuge samples at 3000 rpm for 10 minutes. The resulting supernatant was filtered through filter paper with a pore size of 0.45 μm. According to the change of ammonia nitrogen concentration, the amount of ammonia nitrogen adsorption by the medium is calculated, and the dynamic adsorption curve of ammonia nitrogen by the medium is drawn. The amount of adsorption is calculated:

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

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

Results and discussion

Adsorption isotherm of medium to ammonia nitrogen

The linear equation of Langmuir isotherm adsorption is shown in eq. (2)

$$\frac{1}{q_\mathrm{e}} = \frac{1}{kq_\mathrm{max}C_\mathrm{e}} + \frac{1}{q_\mathrm{max}} \tag{2}$$

where C_e [mgL^{-1}] is the concentration of ammonia nitrogen in the solution for the adsorption equilibrium of each medium, q_e [mgg^{-1}] – the adsorption amount of each medium for the adsorption of ammonia nitrogen equilibrium, q_max [mgg^{-1}] – the single layer adsorption when the equilibrium reaches maximum adsorption capacity, and k [Lmg^{-1}] – the adsorption equilibrium constant.

The results of isothermal adsorption of ammonia nitrogen by various media are shown in figs. 1 and 2. The fitting results of Langmuir equation are shown in tab. 1. The larger the value of k, the more stable the binding between adsorbent and adsorbate [10], and the stronger the adsorption capacity. From the k value, it can be seen that the relationship of the single-layer adsorption capacity of the three media is sand > aluminum powder > iron powder. Because the specific surface area of sand is larger, it provides an additional adsorption point for ammonia nitrogen and increases the adsorption capacity of ammonia nitrogen [11]. Iron powder and aluminium powder have a strong adsorption capacity for ammonia nitrogen, but the data show that the adsorption capacity of aluminium powder is higher than that of iron powder:

– when $R_L = 0$, the ammonia nitrogen adsorption is considered impossible,
– when $0 < R_L < 1$, the ammonia nitrogen adsorption process is feasible [12],
– when $R_L = 1$, the ammonia nitrogen adsorption process is linear, and
– $R_L > 1$ is not conductive to the adsorption of ammonia nitrogen.

The factor-free adsorption strength, R_L, of the three media is $0 < R_L < 1$, indicating that the Langmuir isotherm adsorption process of ammonia nitrogen in the three media is feasible.

As shown in fig. 1, as the initial concentration increases, the adsorption of ammonia nitrogen by the three media also increases. It may be that the higher solution concentration makes the concentration difference between the solution body and the liquid film on the outer surface of the media become larger, thereby increasing the mobility of ammonia nitrogen on the surface of the medium [11]. As shown in fig. 2, the adsorption rate of ammonia nitrogen by the three media showed an increasing trend in the early stage of adsorption. When the initial concentration reached 20~70 mg/L, the adsorption rate of aluminum powder and iron powder for ammonia nitrogen decreased first and then increased. When the ion concentration of sand is 30~70 mg/L, the adsorption rate of ammonia nitrogen first decreases sharply with the increase of ion concentration, and it continues to increase when it reaches a certain concentration. Therefore, the optimal initial concentration conditions for sand, Iron powder and aluminum powder are 30, 20, and 70 mg/L, respectively, and the maximum adsorption rates are 82%, 66%, and 75%, respectively.

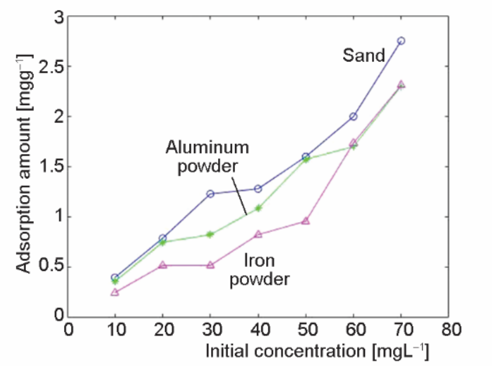

Figure 1. Isothermal adsorption capacity of different media

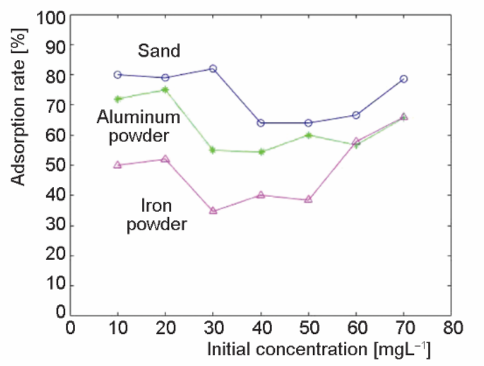

Figure 2. Isothermal adsorption rate of different media

Table 1. Langmuir isotherm adsorption parameters of ammonia nitrogen adsorption in different media

Media	Q_{max} [mgg^{-1}]	k [mgL^{-1}]	R_L	Equation	R^2
Sand	0.6918	90.3418	$0 < R_L < 1$	$y = -0.016x + 1.4455$	0.9077
Iron powder	0.3726	80.6066	$0 < R_L < 1$	$y = -0.0333x + 2.6842$	0.9491
Aluminum powder	0.5850	89.9684	$0 < R_L < 1$	$y = -0.019x + 1.7094$	0.9622

Freundlich isotherm adsorption model

The linear equation of Freundlich isotherm adsorption is:

$$\ln q_e = \ln K_f + \frac{1}{n}\ln C_e \tag{3}$$

where K_f and n are constants, C_e [mgL^{-1}] is the concentration of ammonia nitrogen in the solution when the adsorption equilibrium of each medium, and q_e [mgg^{-1}] – the adsorption amount of each medium when the adsorption of ammonia nitrogen equilibrium.

The adsorption results of the isothermal adsorption of ammonia nitrogen by the three media. The simulation results of Freundlich isotherm adsorption equation are shown in tab. 2. The K_f is related to the affinity of the adsorbent [10]. The larger the K_f, the better the adsorption capacity of the adsorbent to the medium. The K_f values of the three media are sand (0.2698 mg/L), iron powder (0.0753 mg/L), and aluminum powder (0.1963 mg/L). The value of n is between 1 and 10, which is conducive to the ammonia nitrogen adsorption process. The n values of sand, iron powder, and aluminum powder are 1.3026, 1.3827, and 1.3870 respectively, so the three media have a good adsorption effect on ammonia nitrogen. The order of adsorption capacity from high to low is sand > aluminum powder > iron powder, which is consistent with the results of Langmuir model.

Table 2. Freundlich isotherm adsorption parameters for ammonia nitrogen adsorption in different media

Media	K_f [mgL^{-1}]	n	Equation	R^2
Sand	0.2698	1.3026	$y = 0.7677x – 1.31$	0.9064
Iron powder	0.0753	1.3837	$y = 0.7227x – 2.5867$	0.9397
Aluminum powder	0.1963	1.3870	$y = 0.721x – 1.6283$	0.9484

It can be seen from tabs. 1 and 2 that the adsorption effect of sand on ammonia nitrogen is the best, so sand can be used as the adsorption material of ammonia nitrogen. Comparing the correlation coefficient, R^2, of the Langmuir and Freundlich isotherm adsorption models, it can be seen that the adsorption process of ammonia nitrogen by the three media is more in line with the Langmuir isotherm adsorption model, and the single-layer adsorption is mainly used [13].

Effect of pH values on adsorption

The pH value is an important factor affecting the effect of the adsorbent [14]. Not only will it affect the functional groups, charges and active sites on the surface of the adsorbent, but also the degree of ionization of the adsorbate [15]. The results of ammonia adsorption of three media with pH values in the range of 3-6 are shown in fig. 3.

In the range of pH = 3~4, the adsorption capacity of sand soil for ammonia nitrogen gradually decreases. At pH = 4~9, the adsorption capacity of sand soil for ammonia nitrogen increases with the increase of pH value, and reaches saturation state at pH = 9, and at pH = 6, the adsorption capacity of sand soil for ammonia nitrogen reaches the maximum, which is 2.08 mg/g. This is because when the pH value is low, H^+ and NH_4^+ in the solution are competitively adsorbed, and the active site of the sand is occupied by H^+, resulting in a decrease in the amount of ammonia nitrogen adsorption. And the pH value will directly affect the positive and negative charge of the surface of colloidal particles [16]. With the increase of pH value, the surface of the sand has more negative charges, and the ammonia nitrogen has a positive charge, which is conducive to the adsorption of ammonia nitrogen on the sand. When the pH value is 3~5, the adsorption capacity of aluminum powder for ammonia nitrogen gradually increases. In the range of pH = 5~9, the

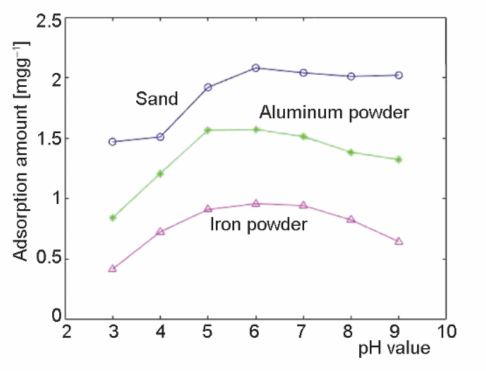

Figure 3. Ammonia nitrogen adsorption capacity of three media at different pH values

adsorption capacity of aluminum powder for ammonia nitrogen gradually decreases. At pH = 5, the adsorption capacity of aluminum powder to ammonia nitrogen reaches the maximum value, which is 1.576 mg/g. This is because as the pH increases, the aluminum powder hydrolyzes in solution, precipitating mainly in the form of Al(OH) [17], resulting in a reduction in the specific surface area of the aluminum powder, which reduces the adsorption of ammonia nitrogen. In the range of pH = 3~6, the adsorption capacity of iron powder for ammonia nitrogen increases with the increase of pH. When the pH is in the range of 6~9, the adsorption capacity of iron powder for ammonia nitrogen decreases. At pH = 6, the adsorption of iron powder to ammonia nitrogen reached the maximum, which was 0.959 mg/g. The main reason is that under acidic environment, it can effectively suppress the deposition of deposits on the surface of iron powder, so that the active sites on the surface of iron powder can be fully exposed, which promotes the adsorption effect of ammonia nitrogen. Sand, iron powder, and aluminum powder have the best adsorption capacity when the pH is 6, 6, and 5, respectively. At pH = 3, the three media have the lowest adsorption capacity for ammonia nitrogen.

Effect of ionic strengths on adsorption

The ammonia nitrogen adsorption capacity of the three media under different ionic strengths is shown in fig. 4. When the ionic strength is 1-20 mmol/L, the ammonia nitrogen adsorption of sand and iron powder is proportional to the ion concentration and increases with the increase of ion concentration. When the ion concentration is 20-100 mmol/L, the amount of ammonia nitrogen adsorption of soil and iron powder decreases with the increase of ion concentration. However, when the sand concentration is 20 mmol/L, the adsorption capacity of ammonia and nitrogen for sand and iron powder reaches the maximum, which is 2.06 mg/g and 0.936 mg/g, respectively. Studies have shown that when there is electrostatic attraction between the adsorbent and the adsorbate, increasing the ionic strength is detrimental to adsorption. When there is electrostatic repulsion between the adsorbent and adsorbate ions, the increase in ion concentration is beneficial to adsorption [18]. It shows that there is electrostatic attraction between sand and ammonia nitrogen and Iron powder and ammonia nitrogen. When the ion concentration is 1-20 mmol/L, the adsorption capacity of aluminum powder to ammonia nitrogen shows a trend of decreasing first and then increasing with the increase of ion concentration. When the ion concentration is 20-100 mmol/L, the adsorption of aluminum powder to ammonia nitrogen. With the increase of ion concentration, it decreases, and the adsorption capacity of aluminum powder to ammonia nitrogen reaches the maximum at 20 mmol/L, which is 1.57 mg/g. It also shows that there is electrostatic attraction between aluminum powder and ammonia nitrogen. Sand, iron powder, and aluminum powder at different ion concentrations have the largest changes in ammonia nitrogen adsorption capacity of 0.81 mg/g, 0.325 mg/g, and 0.341 mg/g, respectively. It shows that the adsorption capacity of sand to ammonia nitrogen is more affected by the ion concentration. The adsorption of ammonia and nitrogen by iron powder is more affected by the ion concentration. In general, the increase in ion concentration

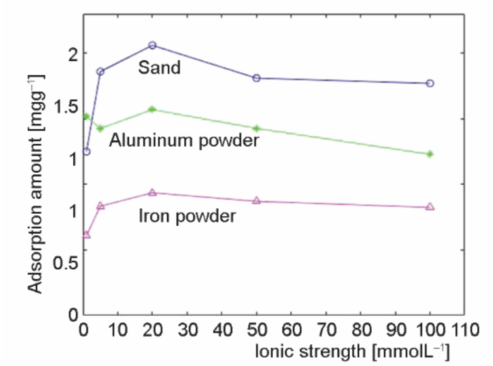

Figure 4. The adsorption capacity of three media to ammonia nitrogen under different ion concentrations

inhibits the adsorption of ammonia nitrogen by the medium, probably because Na^+ and NH_4^+ are both cations, and can ion exchange with substances with ion exchange function on the surface of the medium, so that Na^+ and NH_4^+ have competitive adsorption [19]. Therefore, when the ion concentration is at a low level (1-20 mmol/L), the increase in ion concentration is beneficial to the adsorption of ammonia nitrogen by the medium.

Experimental results of adsorption kinetics of different types of media for ammonia nitrogen

First-order kinetic model

The meanings of the first order kinetic equations and related parameters are as follows:

$$\ln(q_\mathrm{e} - q_t) = \ln q_\mathrm{e} - k_1 t \tag{4}$$

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

The quasi-secondary kinetic model

The meanings of the second-order kinetic equations and related parameters are:

$$\frac{t}{q_t} = \frac{1}{k_2 q^2} + \frac{t}{q_\mathrm{e}} \tag{5}$$

where q_e [mgg^{-1}] is the adsorption amount of each medium when ammonia nitrogen adsorption is in equilibrium, q_t [mgg^{-1}] – the adsorption amount of the medium to ammonia nitrogen at time t, and k_2 [min^{-1}] – the quasi-second-order kinetic rate constant.

The adsorption kinetics of sand, iron powder, and aluminum powder are shown in fig. 5. Within 1~6 hours, the adsorption capacity of aluminum powder for ammonia nitrogen increases with time, while sand shows a downward trend at 5-6 hours. The iron powder sorption capacity of ammonia nitrogen gradually increased with the extension of time within 1-4 hours. The adsorption capacity of iron powder ammonia nitrogen showed a decreasing trend within the range of 4-6 hours. In the initial stage of adsorption, there are a large number of active sites on the surface of the medium, the concentration of adsorbate at the interface of the medium and the solution is large, and the gradient of the adsorption power is large, which is conducive to adsorption [13]. As the adsorption time progresses, the adsorption sites on the surface of the medium are consumed, resulting in a decrease in the amount of ammonia nitrogen adsorbed by the medium.

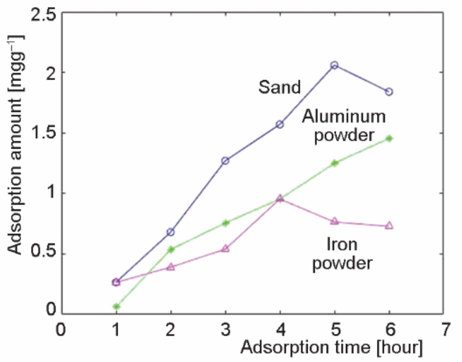

Figure 5. Distribution of ammonia nitrogen adsorption results by bioretentions over time

In order to make the adsorption characteristics of ammonia nitrogen for sand, iron powder, and aluminum powder more intuitive, the following first-order kinetic equation and the quasi-secondary kinetic model are adopted for the adsorption of ammonia nitrogen on sand, iron powder and aluminum powder perform fitting.

The results are shown in tab. 3. In the first-order kinetic model, the R^2 of the three media are sand (0.9912), iron powder (0.9209), and aluminum powder (0.9413). The R^2 in the quasi-secondary kinetic model is sand (0.6495), iron powder (0.5484), aluminum powder (0.5386). From the correlation coefficients, it can be known that the adsorption process of ammonia nitrogen by these three media is more suitable for fitting with the first-order kinetic model, which indicates that the adsorption of ammonia nitrogen by the three media is mainly physical adsorption.

Table 3. Calculation results of the first-order and second-order kinetic models at 298 K

Media	First-order kinetic model		Quasi-secondary kinetic model			
	R^2	q_e [mgg^{-1}]	k_1 [min^{-1}]	R^2	q_e [mgg^{-1}]	k_2 [10^{-3}min^{-1}]
Sand	0.9912	2.06	0.0073	0.6495	3.836	0.00077
Iron powder	0.9209	0.954	0.0049	0.5484	1.3827	2.871
Aluminum powder	0.9413	0.0074	0.4267	0.5368	11.99	0.0316

Conclusions

In this paper, the kinetics of ammonia nitrogen adsorption is studied, and the results are as follows.

- The first-order kinetic model is more suitable than the quasi-secondary kinetic model for the ammonia nitrogen adsorption kinetic model of sand, iron powder, and aluminum powder. Therefore, the adsorption of ammonia nitrogen by the three media is mainly physical adsorption. The adsorption isotherms of ammonia nitrogen for the three media are consistent with Langmuir's adsorption equation. The results show that the relationship between the adsorption capacity of the three media for ammonia nitrogen is sand > aluminum powder > iron powder.
- Isothermal adsorption experiments show that with the increase of the initial concentration, the adsorption of ammonia nitrogen by sand, iron powder, and aluminum powder continues to increase. When the concentration of ammonia nitrogen in the solution reaches a certain level, the adsorption rate of the medium to ammonia nitrogen begins to decrease. The minimum removal rates of sand, iron powder and aluminum powder were 64%, 34.7%, and 54.3%, respectively.
- The adsorption performance of the three media for ammonia nitrogen is greatly affected by the pH value. The optimal pH for ammonia nitrogen adsorption by sand, iron powder and aluminum powder is 6, 6, and 5, respectively.
- The change of ion concentration has a greater effect on the adsorption of ammonia nitrogen on sand. In terms of metal, the adsorption performance of aluminum powder on ammonia nitrogen is more affected by the ion concentration than that of iron powder. The optimum ion concentration of sand, iron powder and aluminum powder for ammonia nitrogen adsorption is 20 mmol/L.

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