



Adsorption Performance of Four Substrates in Constructed Wetlands for Nitrogen and Phosphorus Removal

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ABSTRACT

In this research, nitrogen and phosphorus adsorption characteristics of four substrates, including lava, activated carbon, furnace slag and spongy iron, were studied through basic experiments. Meanwhile, adsorption kinetics, effects of different pH on adsorption performance, sequencing batch adsorption isotherm experiments and saturation analysis of substrates experiments were analysed. Moreover, the possible secondary pollution risks after four substrates saturation adsorption were discussed. The results showed that Freundlich and Langmuir models were favourably fit for the variance on nitrogen and phosphorus adsorption characteristics of substrates. The order of ammonia nitrogen adsorption capacity for four substrates was activated carbon>furnace slag>lava>spongy iron. In addition, judging from ammonia nitrogen desorption rates, the capacity of releasing ammonia nitrogen for four substrates increased as an order of spongy iron>lava>furnace slag>activated carbon. Besides, the order of phosphorus adsorption capacity for four substrates was activated carbon>spongy iron>furnace slag>lava. And the capacity of releasing phosphorus after phosphorus desorption for four substrates increased as an order of lava>spongy iron>furnace slag>activated carbon. In summary, after adsorption and desorption properties of four substrates were analysed, it could be concluded that activated carbon was a more favourable filler for constructed wetlands.

INTRODUCTION

In the 1970s, constructed wetlands began to be used for sewage treatment, and the wastewater treatment systems had attracted great attention because of environmental, economic and social benefits (Bubba et al. 2003, Arias et al. 2001, Brooks et al. 2000, Zhu et al. 2011). Advantages of constructed wetlands could be expressed with low construction and operating costs, good water quality, simple operation and beautifying the environment (Zhao et al. 2009). Meanwhile, constructed wetlands were a suitable emerging technology for wastewater treatment, especially to address the water quality problem in rural areas and small towns, which had a broad development prospects (Summers et al. 1993, Greenway. 2003). Raising a virtuous cycle of pollutants was the premise to prevent secondary pollution of the environment and resources, and maximize the potential of constructed wetlands (Kaya et al. 2009, Precup et al. 2011, Liu et al. 2012, Vohla et al. 2011). In addition, constructed wetlands could also be used as a conventional sewage treatment technology as an effective alternative to achieve the best effect of sewage treatment (Prochaska et al. 2006, Drizo

et al. 2006). At present, constructed wetlands have been widely used in domestic sewage, industrial effluent, farm wastewater, mining and oil extraction wastewater treatment, in order to control the eutrophication problem and ecological restoration of polluted water (Wen et al. 2010, Fu et al. 2009). Therefore, a new option was provided for sewage nitrogen and phosphorus removal in constructed wetland ecosystems (Tao et al. 2009). Besides, constructed wetland was an artificial construction and supervision similar to the marsh flat, and its basic components include substrates, plants and microorganisms (Guo et al. 2010, Yang et al. 2010). The course of wastewater treatment in constructed wetlands involved several aspects including the use of substrates, plants and microorganisms with physical, chemical and biochemical processes, such as synergy through filtration, ion exchange, adsorption, precipitation, soil absorption, etc.

Constructed wetland substrates are also called waddings, and traditional substrates of constructed wetlands include soil, sand, gravel, etc. In recent years, crushed rock, zeolite, activated carbon and ceramics also began to be widely used

in constructed wetlands. Substrate is the main medium for removal of pollutants in constructed wetland, which could obviously remove nitrogen, phosphorus and other nutrients in the wastewater through physical and chemical methods. In this research, the adsorption characteristics of lava, activated carbon, furnace slag and spongy iron for ammonia nitrogen and phosphorus are discussed, in order to provide data for further engineering application foundation on constructed wetlands.

MATERIALS AND METHODS

Various Characteristics of Substrates

Adsorption kinetics of matrix experiment: In the experiments, the four substrates were weighed of 20g and put into 250mL flasks, respectively. Then the NH_4Cl and KH_2PO_4 were added into prepared solution with ammonia nitrogen concentration of 30mg/L and phosphorus concentration 15mg/L. Under conditions of 25°C and 129r/min, oscillation adsorption was applied in oscillator. The nitrogen and phosphorus concentrations were sampled at different time after 0.45 μm membrane filter, and nitrogen and phosphorus adsorption kinetics of the substrates were studied.

Effects of different pH on adsorption performance of matrix experiment: Under the aforementioned experiment condition, the NH_4Cl and KH_2PO_4 were added into prepared solution with ammonia nitrogen concentration of 30mg/L and phosphorus concentration of 9.5mg/L. At the same time, HCl and NaOH were applied to adjust pH to 6, 7, 8 and 10, respectively. Under the conditions of 25°C and 129r/min, oscillation adsorption was applied in oscillator, and sampling at different times after 0.45 μm membrane filter, nitrogen and phosphorus concentrations were analysed and isothermal adsorption curves were drawn.

Sequencing batch adsorption isotherm experiments: Under the aforementioned experimental conditions, the NH_4Cl and KH_2PO_4 were added into prepared ammonia nitrogen solutions with different concentrations of 200mL, namely 13mg/L, 23mg/L, 26mg/L and 33mg/L, at the same time, phosphorus solution of 200mL, namely 6 mg/L, 8 mg/L, 15 mg/L and 18 mg/L, respectively. Under the conditions of 25°C and 129r/min, oscillation adsorption was applied in oscillator for 48 hours, and sampling at different times after 0.45 μm membrane filtration.

Saturation analysis of matrix experiment: Under the aforementioned experiment condition, four 250mL flasks were applied to substrates adsorption, and every flask was put into 10g saturated substrates and deionized water 200mL. Under the conditions of 25°C and 129r/min, oscillation adsorption was applied in oscillator for 24 hours, and sam-

pling at different times after 0.45 μm membrane filtration. Meanwhile, removal effects of nitrogen and phosphorus concentration were analysed.

RESULTS AND DISCUSSION

Adsorption kinetics of substrates:

As is shown in Fig. 1, the order of phosphorus adsorption capacities for four substrates was activated carbon > spongy iron > furnace slag > lava. Phosphorus adsorption of sponge iron during 100 minutes for micro-polluted water was fast, and low phosphorus content remained with the increase of time. Adsorption saturation characteristics of sponge iron were exhibited through the degradation of phosphorus in micro-polluted water, thus a small amount of phosphorus content in micro-polluted water increased. Activated carbon and furnace slag could continuously retain phosphorus absorption in a long time, and phosphorus content attained low value in micro-polluted water. However, phosphorus adsorption of volcanic rocks had no significant effect with adsorption time increased.

As is shown in Fig. 2, the order of adsorption capacity for four substrates on ammonia nitrogen was activated carbon > furnace slag > lava > spongy iron. Adsorption of ammonia nitrogen for activated carbon and furnace slag showed excellent effect, and adsorption capacity had little difference. Meanwhile, activated carbon and furnace slag exhibited favourable adsorption characteristics for nitrogen in a short period of time. But the nitrogen content in the solution did not increase continuously with the increase of time, and slowed down once again until it reached saturation point. The absorption time of sponge iron and lava was 200 minutes for nitrogen from micro-polluted water, and nitrogen adsorption effects were unsatisfactory with the increasing of adsorption time. In addition, compared with lava, the activated carbon and furnace slag for nitrogen and phosphorus exhibited better adsorption performance. Besides, the reason of substrates absorption properties was surface tension, and there was attractiveness to the outside on a solid surface and lateral surface adsorption of other particles. Activated carbon was a porous material with a lot of micro-pores, which induced that the surface area was large, and had more effective absorption for nitrogen and phosphorus.

Effects of different pH on the adsorption property of substrates analysis: Adsorption characteristics of nitrogen in different pH: As is shown in Fig. 3, lava showed a relatively clear adsorption effect under the condition of pH < 8. Nitrogen adsorption capacity of lava increased with the pH values when the solution was acidic, and nitrogen adsorption of lava was lower, and it was hard to be adsorbed

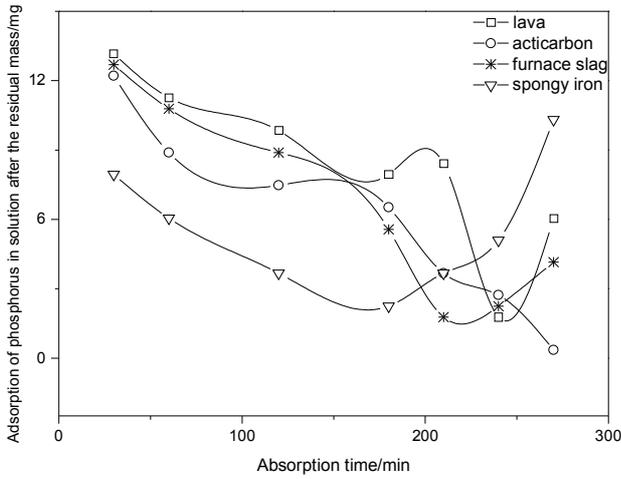


Fig. 1: The effect of phosphorus adsorption curves on different substrates.

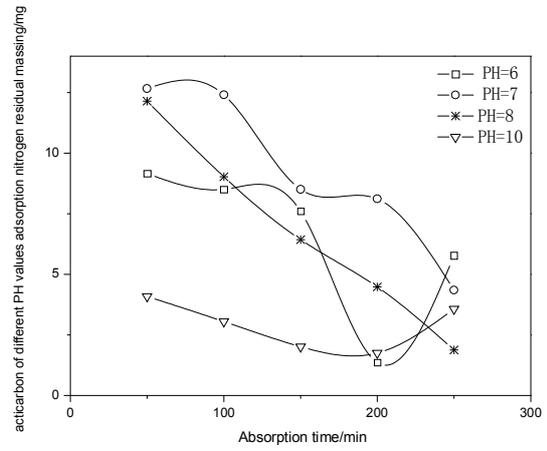


Fig. 4: The effect of ammonia nitrogen adsorption with activated carbon in different pH

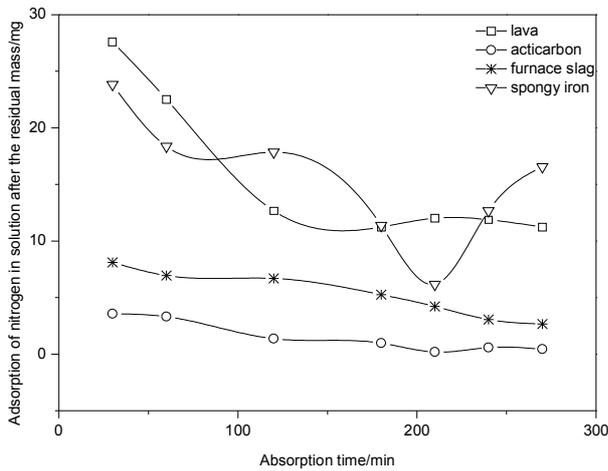


Fig. 2: The effect of nitrogen adsorption curves on different substrates.

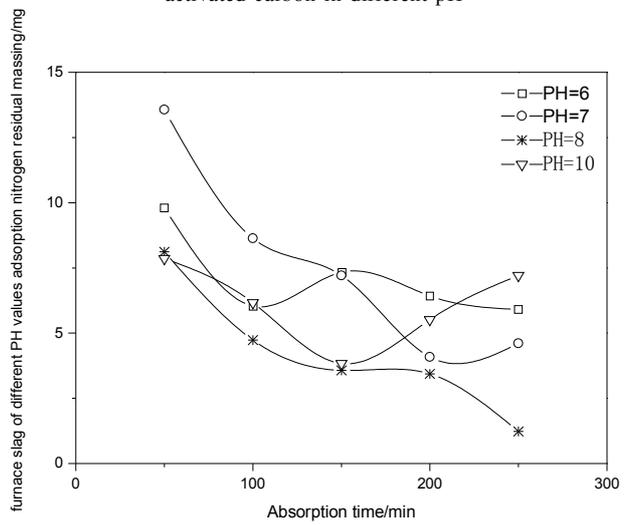


Fig. 5: The effect of ammonia nitrogen adsorption with furnace slag in different pH.

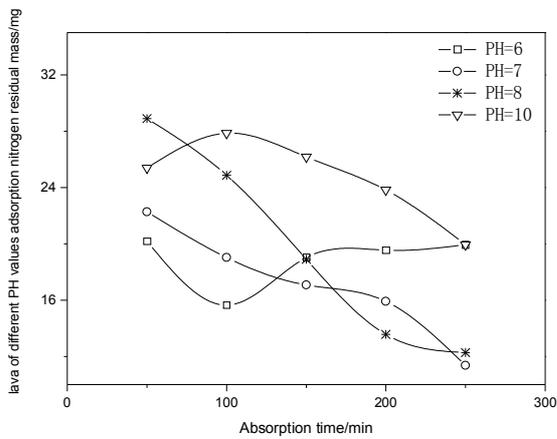


Fig. 3: The effect of ammonia nitrogen adsorption with lava in different pH.

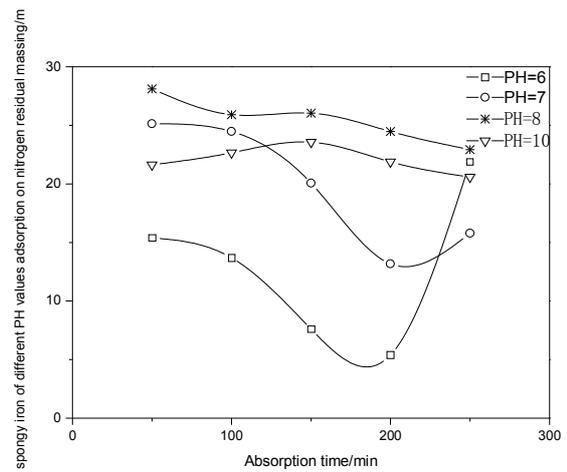


Fig. 6: The effect of ammonia nitrogen adsorption with spongy iron in different pH.

Table 1: Characteristics of substrates used in the experiments.

Item/Substrates	Trace elements	Chemical stability	Specific surface area	Porosity (%)	Packing density (g/cm ³)
Lava	Na, Mg, Al, Si, Ca, Ti, Mn, Mu, P, Fe, Co	General	Large	77.0	0.74
Acticarbon	C, O, H, S, N, Cl	Excellent	Large	27.5	0.58
Furnace slag	SiO ₂ , Al ₂ O ₃ , CaO, MgO, MnO, Fe ₂ O ₃	Good	General	31.8	1.8
Spongy iron	C, Fe, Mn, Ni, Cr, CaO, MgO	Good	Very big	40	1.7

Table 2: Relative parameters of ammonia nitrogen adsorption isotherm equation for the four substrates (25°C).

Substrates	Freundlich's adsorption formula				Langmuir's adsorption formula		
	K	1/n	R(n=4)	G ₀ (mg/g)	A	R(n=4)	MBC(mg/g)
Lava	0.0550	0.51769	0.90504	0.5435	0.0296	0.95937	18.36149
Acticarbon	1.038944	0.18426	0.98809	1.173544	0.05411	0.97973	21.6833
Furnace slag	0.999913	0.38582	0.91689	1.030454	1.06356	0.92341	0.968869
Spongy iron	0.73570	2.14771	0.98864	0.005574	12.6904	0.994987	0.000492

Table 3: Relative parameters of phosphorus adsorption isotherm equation for the four substrates (25°C).

Substrates	Freundlich's adsorption formula				Langmuir's adsorption formula		
	K	1/n	R(n=4)	G ₀ (mg/g)	A	R(n=4)	MBC(mg/g)
Lava	0.0209	0.6870	0.98889	0.23120	0.19325	0.99679	1.196378
Acticarbon	0.036	0.27758	0.98691	0.46115	0.073305	0.958645	6.29077
Furnace slag	0.057122	0.41331	0.92459	0.50124	0.122995	0.971289	4.07527
Spongy iron	0.000424	0.63662	0.98356	0.15152	3.43997	0.998299	0.04405

Table 4: Ratio of liberation and adsorption of ammonia and phosphorus in substrates saturated with ammonia and phosphorus.

Substrates	Lava	Acticarbon	Spongy iron	Furnace slag
Maximal adsorptive capacity of ammonia nitrogen in theory (mg/g)	0.5435	1.73544	0.005574	1.030454
Maximal liberation capacity of ammonia nitrogen (mg/g)	0.233748	0.025976	0.301841	0.168844
Liberation percentage of ammonia nitrogen (%)	7.569365	2.612915	6.481142	4.478944
Ammonia nitrogen concentration after liberation (mg/L)	26.9371	4.2081	20.691	11.7419
Maximal adsorptive capacity of phosphorus in theory (mg/g)	0.23120	0.461145	0.15152	0.501238
Maximal liberation capacity of phosphorus (mg/g)	0.485895	0.0378265	0.22426	0.074753
Liberation percentage of phosphorus (%)	5.040669	1.83969	2.86492	2.642995
Phosphorus concentration after liberation (mg/L)	6.61455	2.28745	4.451	5.0251

in solution. In general, the nitrogen adsorption was not satisfied for lava under different pH values.

As is shown in Fig. 4, adsorption of nitrogen by activated carbon was most prominent when the pH was more than 8. In particular, nitrogen in solution dropped to a very low value when pH was 10. When solution was neutral or acidic, less nitrogen content from micro-polluted water was adsorbed by activated carbon, but activated carbon exhibited a favourable adsorption effect for nitrogen.

As shown in Fig. 5, effect of pH values on the slag adsorption of nitrogen may be neglected. Nitrogen adsorption in alkaline solution was relatively more obvious, but fur-

nace slag in nitrogen adsorption was affected under different pH values.

As shown in Fig. 6, sponge iron exhibited the larger amount of nitrogen adsorption capacities in solution and fast adsorption speed when pH was less than 6. When pH was neutral or alkaline, sponge iron had a low nitrogen adsorption characteristic, so the content of nitrogen in micro-polluted water has not changed. Compared with activated carbon, sponge iron exhibited slightly inferior adsorption effect for nitrogen.

As are shown in Figs. 3-6, because the electrode could produce Fe²⁺ and further oxidation of Fe³⁺ in acidic condi-

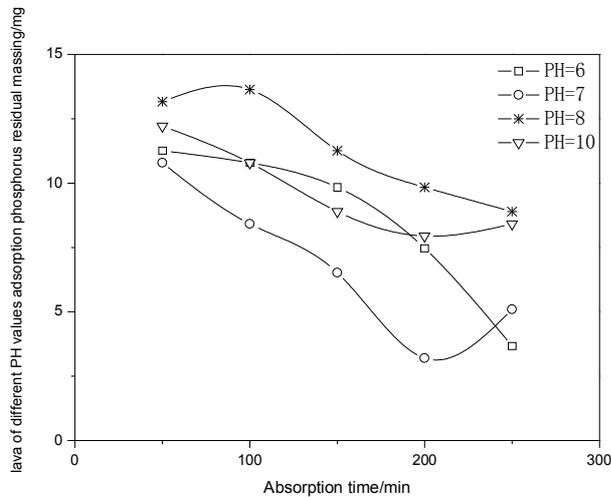


Fig. 7: The effect of phosphorus adsorption with lava in different pH.

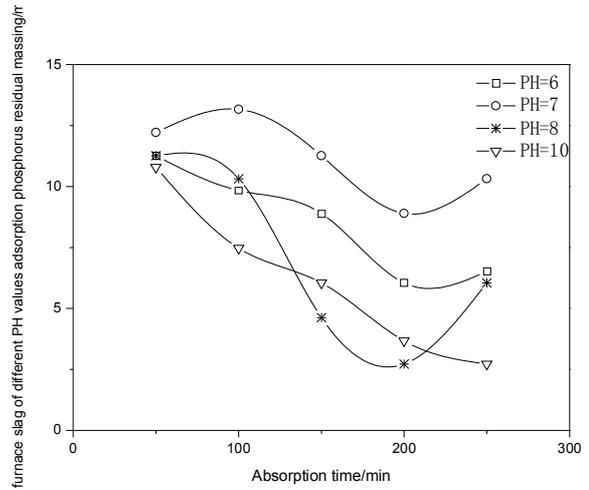


Fig. 9: The effect of phosphorus adsorption with furnace slag in different pH.

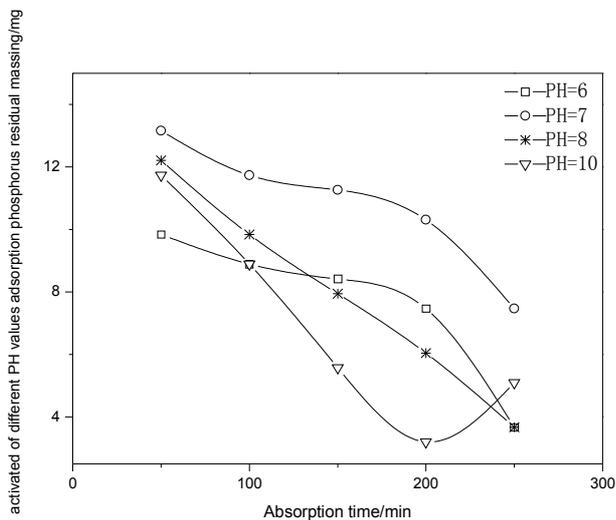


Fig. 8: The effect of phosphorus adsorption with activated carbon in different pH.

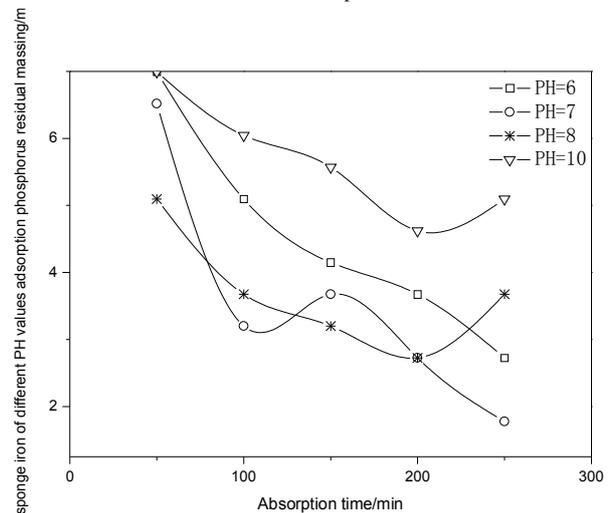
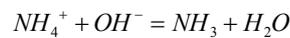


Fig. 10: The effect of phosphorus adsorption with spongy iron in different pH.

tion, when the pH was adjusted to alkaline and oxygen is present, it could come into being flocculent precipitate of $Fe(OH)_2$ and $Fe(OH)_3$. In addition, H^+ had a characteristic with exchange-adsorption competition of NH_4^+ , ammonia nitrogen removal rate was lower under acidic conditions. Apart from adsorption effect for ammonia nitrogen of substrates, ammonia nitrogen volatilization also was a factor while pH values in solution affected ammonia volatilization. Ammonia volatilization was almost nonexistent when pH was 7, but in contrast, the ratio of ammonium ions and hydroxyl ions was 1:1 and volatilization of ammonia removal started to become significant when the pH was 10. Its reaction equation is:



Meanwhile, compared with sponge iron and lava, activated carbon and furnace slag exhibited favourable ammonia nitrogen adsorption capacities.

Adsorption characteristics of phosphorus in different pH:

As is shown in Fig. 7, phosphorus adsorption of lava was changed lightly under different pH. Namely, lava exhibited unsatisfactory phosphorus adsorption effects when the reaction solution was acidic. Fig. 8 shows the adsorption effect of activated carbon for phosphorus under different pH. When the solution was alkaline, activated carbon adsorption effect of phosphorus in micro-polluted water increased

with the time. As is shown in Fig. 9, furnace slag exhibited a certain adsorption capacities under different pH values. Adsorption effect was better in alkaline solution than acidic. Therefore, it showed good adsorption under alkaline conditions. Meanwhile, Fig. 10 shows a large amount of phosphorus adsorption capacities with sponge iron. When the solution was acidic or neutral, phosphorus adsorption of sponge iron was better and the adsorption capacity increased with the time.

As is shown in Figs. 7-10, phosphorus adsorption effects of activated carbon and sponge iron were better than the lava and furnace slag. The studies have shown that Al^{3+} , Fe^{2+} , Ca^{2+} , Mg^{2+} , Mn^{2+} are generated by precipitation or adsorption. With the increase of pH value, solution of iron and aluminium ions with phosphate-binding capacity would diminish, and iron aluminium salts resulted instability of colloidal precipitation or dissolution. Iron and aluminium salts were the main products in the lava and sponge iron, so adsorption properties of phosphorus diminished with the increase of pH. In the $H_2PO_4^-$ solution, $H_2PO_4^-$ turned into HPO_4^{2-} as pH increased, but HPO_4^{2-} , Ca and Mg could generate precipitation. Activated carbon and sponge iron decreased as pH increased, and the phosphorus adsorption speed was gradually reduced. In addition, calcium and iron aluminium are transferred into substrates, and they had a strong capability of sewage purification. A lot of silica substrates possessed the characteristics of poor purification ability. Beside, studies had shown that calcium content was the main factor to affect the phosphorus removal ability. High calcium content contributed into insoluble calcium phosphate precipitate in alkaline water. At the same time, iron and aluminium content in acidic wastewater was more important for precipitation.

Substrates on nitrogen and phosphorus adsorption isotherms experiment: Under the condition of constant temperature, Freundlich adsorption equation is commonly used to represent the solid surface adsorption and adsorption equilibrium concentration in liquids, and Langmuir adsorption equation was determined to the theoretical maximum adsorption capacity.

Freundlich adsorption equation is:

$$\lg G = \lg K + \frac{1}{n} \lg C$$

Where, G represents amount of adsorption equilibrium of adsorbent per unit mass; C represents adsorbent in adsorption equilibrium concentrations; and K and n are constants.

In the Freundlich equation, parameters of adsorbate and adsorbent express adsorption capability, $1/n$ is the function of adhesion. For determining the C and $1/n$, K is the amount

of adsorption of the direct response, so stronger and larger k values indicate higher adsorption capacity. For determining the K and C, $1/n$ is the amount of adsorption of the direct response. Adsorption capacity has nothing to do with C when $1/n$ is small. And adsorption isotherm is close to horizontal line and G is almost a constant at this time. If $1/n$ is large adsorption force, G and C changes significantly. Meanwhile, $1/n$ could be roughly the reaction substrates on the strength of nitrogen and phosphorus adsorption. It was generally considered that adsorbate is easily adsorbed when $1/n$ is less than 0.5, in contrast, adsorbate was difficult to be adsorbed when $1/n$ is more than 2.

$$\text{Langmuir adsorption equation: } \frac{1}{G} = \frac{1}{G_0} + \frac{A}{G_0} \frac{1}{C}$$

Where, G_0 represents amount of the theoretical maximum adsorption of adsorbent; C represents adsorbent in adsorption equilibrium concentrations; G represents the quality of adsorbent in adsorption equilibrium; A is the constant.

In the Langmuir equation, G_0 expresses the number of adsorbance, and adsorption increases with G_0 . $1/A$ shows the nitrogen complexing ability and maximum adsorption amount of substrates. According to the experimental results, Langmuir and Freundlich equations were used to fit the adsorption rules. Adsorption substrates adsorption isotherm parameters of ammonia nitrogen are shown at Table 2 and phosphorus parameters are shown in Table 3.

As given in Table 2, Freundlich and Langmuir adsorption models of the four substrates for ammonia nitrogen were fitted, which showed good correlations. The order of K-values for the four substrates was activated carbon > furnace slag > sponge iron > lava. Sponge iron k-value was larger than 2, so sponge iron has a poor adsorption effect for ammonia nitrogen. When $1/n$ values of activated carbon and furnace slag were less than 0.5, they could easily adsorb the ammonia nitrogen. In addition, the order of G_0 values for four substrates were activated carbon > furnace slag > lava > sponge iron. And the order of MBC values were followed by activated carbon > lava > furnace slag > sponge iron. Besides, k, G_0 and MBC values of the activated carbon were the maximum, and G_0 , MBC values were far more than other three substrates. Thus, activated carbon had larger adsorption and buffer capacities and the adsorbance and buffer capacity of furnace slag and lava were smaller, sponge iron was smallest. Therefore, sponge iron should not be applied as a single packing material when building the constructed wetlands with high loading of ammonia nitrogen. However, activated carbon could be considered as favourable single filler or added into other mixed substrates.

As is indicated in Table 3, Freundlich and Langmuir

adsorption models of four substrates about phosphorus were fitted, which also showed good correlations. The order of K -values for four substrates was furnace slag > activated carbon > lava > sponge iron. Only activated carbon and furnace slag' $1/n$ values were less than 0.5. Adsorption effect of activated carbon and furnace slag for phosphorus was extremely easy, but sponge iron and lava were difficult. The order of G_0 values for four substrates was furnace slag > activated carbon > lava > sponge iron. Meanwhile, the order of MBC values was followed by activated carbon > furnace slag > lava > sponge iron. In addition, k , G_0 and MBC values of activated carbon were the maximum. Besides, four substrates have good adsorbance and buffer capacity and the adsorbance and buffer capacity of sponge iron was smallest. But the G_0 and MBC values remained low. In general, substrates were considered as final destination for phosphorus, which entered the wetlands. Thus, appropriate substrates were chosen as fillers for phosphorus adsorption in constructed wetlands. These four substrates couldn't simply act as fillers and other substrates should be found for phosphorus adsorption.

Substrates saturation characteristics of secondary pollution risk assessment: As shown in Table 4, ammonia nitrogen desorption of sponge iron was maximum, next is the furnace slag and lava, and activated carbon was much less than the other three substrates. Activated carbon was also the smallest from the analysis in terms of the percentage, but its adsorption capacity was large. Therefore, activated carbon was favourable substrate for ammonia nitrogen removal in constructed wetlands. Largest desorption of phosphorus was lava and the smallest was activated carbon. In terms of percentage, lava was the largest and the activated carbon was smallest. Under the actual operation for intermittent treatment of domestic wastewater in subsurface constructed wetlands, the hydraulic retention time was often longer than saturated substrate reached the analytical balance. Meanwhile, Table 1 and Table 3 concluded that activated carbon adsorption for ammonia nitrogen with as filler demanded the national wastewater discharge standards, and other three substrates exceeded the relevant national standards. For effluent concentrations of phosphorus, four substrates were not up to standards and far exceeded the national standard. There was a risk of secondary pollution, so we should be careful to choose fillers.

CONCLUSIONS

Substrate dynamics experiments were applied to the substrates performance for ammonia nitrogen and phosphorus adsorption. The results showed that activated carbon adsorption rate for ammonia nitrogen and phosphorus was larger than the other three substrates. Meanwhile, phosphorus adsorption by sponge iron was the best, and furnace slag

and lava was small. Ammonia nitrogen adsorption by furnace slag was relatively larger than sponge iron and lava. For the influence of pH, adsorption capacity of four substrates on ammonia nitrogen and phosphorus were relatively significant. In addition, activated carbon and furnace slag were easy for adsorption of ammonia nitrogen and phosphorus under alkaline conditions, and sponge iron was easy for removal of ammonia nitrogen and phosphorus under neutral and acidic conditions. Adsorption isotherms of the four substrates models were fitted and adsorption of activated carbon and furnace slag on ammonia nitrogen had a larger capacity than lava and sponge iron. Phosphorus adsorption of activated carbon and sponge iron had a larger capacity than lava and furnace slag. These experiments show that the lava, activated carbon, furnace slag and spongy iron had good adsorption performance, so the four substrates could be concluded as favourable filter materials in constructed wetlands.

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