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Adsorption characteristics of recycled construction waste particles for ammonia-nitrogen in rainfall runoff

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Abstract: In order to explore the removal method of ammonia nitrogen in water and solve the disposal problem of construction waste. After the construction waste is crushed, the particles with different sizes (recycled particles for short) are screened out for ammonia nitrogen adsorption in simulated wastewater.SEM,EDS and BET were used to characterize the recycled particles, and the effects of different external factors on adsorption efficiency and ammonia nitrogen removal rate under dynamic environment were investigated. The results show that: The surface of the recycled particles was rough and had a large number of pores. The adsorption process follows the pseudo-second kinetics and Langmuir isotherm adsorption model; the maximum adsorption capacity of the recycled particles of 0-2mm, 2-3mm, 3-5mm and 5-10mm were 0.1169mg/g, 0.1213mg/g, 0.1200mg/g and 0.1182 mg/g, respectively; the optimal pH was 6-7; Na⁺ had a slight effect on ammonia nitrogen adsorption.In the static adsorption experiment, the large size recycled particles have a higher adsorption capacity, while in the dynamic adsorption experiment, the small recycled particles have a higher adsorption capacity.

1. Introduction

Ammonia nitrogen pollution is very common in our country. At present, the main removal ways for ammonia nitrogen are blow off, adsorption, biological treatment, membrane treatment, etc^[1].The technology of membrane treatment and microbial remediation have some imperfection, such as high cost, difficult operation and energy consumption. The adsorption method has the advantages of convenient operation and ideal removal effect and is widely used. Currently adsorbents are zeolite, biochar. Although the currently used zeolite, biochar or modified materials have good adsorption capacity, but they still have some shortages, Such as the cost, the production of biochar is energy consuming, and it sinks to the bottom of the water, which may cause siltation and difficult to be cleaned up. A cheaper and easier-to-manufacture adsorbent needs to explore.

With the acceleration of the urbanization, a large amount of construction and demolition waste (CDW) has been generated. On the one hand, CDW is used as recycled construction material because of its low cost and stability. It is used for roadbed and concrete^[2]. On the other hand, because of its porosity, high specific surface area, adsorption performance, the research on its use as adsorbent is also increasing.

In this experiment, recycled particles of different sizes were applied to the ammonia nitrogen adsorption experiment, and the basic properties of the recycled particles were studied. The adsorption

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characteristics of ammonia nitrogen, the effects of different solution pH, coexisting ion concentrations, and different particle sizes on the adsorption performance of ammonia nitrogen, and the adsorption performance of recycled particles under spraying conditions have been studied and discussed, which provides resources for the utilization of recycled particles theory support.

2. Materials and methods

2.1. Material preparation

Recycled particles are derived from the CDW generated from Shanghai downtown. The CDW is crushed, then sieved into 0-2mm, 2-3mm, 3-5mm, 5-10mm, 4 kinds of particles with different particle sizes, first rinse the particles with tap water and soak them in tap water for 2h, remove the impurity floating on the water during the soaking process and pick up the particles and soak in distilled water for 1h, then place the particles in the oven. The particles is heated at 105°C to constant weight are referred to as recycled particles.

Ammonia nitrogen sewage is prepared by weighing 3.819g NH₄Cl and dissolving it in water to a constant volume of 1L. This solution contains 1g/L ammonia, which is used as mother liquor to prepare ammonia nitrogen sewage with corresponding concentration. All drugs in the experiment are of analytical grade.

2.2. Experimental method

2.2.1. Characterization of recycled particles and sample analysis. Use the S-3400N Scanning Electron Microscope (SEM) to observe the apparent morphology of construction waste recycled particles and combine with an energy spectrometer (EDS) to determine the content of each element in the material; The specific surface area of recycled particles was determined by the Brunauer-Emmett-Teller(BET) methed using the adsorption isotherm data at -196°C using Micromeritics ASAP 2020 V3. 01 E physical adsorption analyzer.

In the experiment, the ammonia nitrogen concentration was determined by Nessler's reagent spectrophotometry, and the adsorption rate and adsorption capacity were calculated by formulas (1) and (2):

$$\eta = \frac{C_0 - C_t}{C_0} \times 100\%$$
(1)
$$q_e = \frac{(C_0 - C_t)V}{m}$$
(2)

Where η is the removal rate (%); C₀ is the initial concentration of ammonia nitrogen (mg/L); C_t is the concentration of ammonia nitrogen in the solution at t (mg/L); q_e is the adsorption capacity (mg/g); V is the volume of solution (L); m is the weight of recycled particles (g).

2.2.2. *Kinetic adsorption experiment*. Weigh 100g of recycled particles of four different particle sizes into a 250mL bottles, add 150mL of ammonia nitrogen solution with a mass concentration of 10mg/L to the bottles, and them were shaken at 25°C of 150r/min. At the 1th, 5th, 10th, 20th, 30th, 40th, 60th, 90th, 120th, 180th, 240th, 300th min 4mL sample were taken and through 0.45µm filter membrane to determine the ammonia nitrogen concentration.

2.2.3. Isothermal adsorption experiment. Weigh 1g of 4 kinds of recycled particles with different particle sizes into 150mL bottles, and add 30mL of ammonia nitrogen solution with concentration of 2mg/L, 3mg/L, 4mg/L, 5mg/L, 6mg/L, and them were shaken at 25°C of 150r/min. After 24h, the solution was taken with a needle syringe and through a 0.45 μ m filter membrane to determine the equilibrium concentration of the solution.

2.2.4. The influence of solution pH.Weigh 100g of 4 kinds of recycled particles with different particle diameters into a 250mL bottle. Add 150mL of solution with pH of 2, 4, 6, 7, 9and an ammonia nitrogen mass concentration of 10mg/L into the bottles. After being plugged and sealed, the bottles were shaken at 25°C of 150r/min., and after 24h, the liquid was taken through a $0.45\mu m$ filter membrane to determine the ammonia nitrogen content in the solution.

2.2.5. The influence of ionic strength. Weigh 100g of 4 kinds of recycled particles with different particle sizes into a 250mL bottle respectively, add 150mL NH₄Cl+NaCl solution, NaCl molar concentration is 0.001mol/L, 0.0025mol/L, 0.005mol/L, 0.01mol/L, 0.05 mol/L respectively, the ammonia nitrogen concentration is 10mg/L. The bottles were shaken at 25°C of 150r/min. After 24h, the solution was taken through a 0.45 μ m filter membrane to determine the ammonia nitrogen content in the solution.

2.2.6. Dynamic adsorption test. The dynamic adsorption experiment was carried out using the device shown in figure 1. The water storage tank was equipped with ammonia nitrogen waste water with a mass concentration of 10 mg/L, and the water flow at the spray outlet was maintained at 70 mL/min. Add 2kg of recycled particles of different particle sizes to the 4 adsorption columns, and lay a layer of qualitative filter paper at the bottom of the column to prevent the loss of recycled particles. After the spraying started, the water discharge time of each adsorption column was recorded. At the 5th, 10th, 15th, 30th, 45th, 60th, 90th, and 120th minutes after the start of the water discharge, 50 mL of water samples were taken from the water outlet to determine the ammonia nitrogen concentration.



Figure 1. dynamic adsorption device.

3.Results and discussion

3.1. Properties of recycled granular materials

Three main types of recycled particles: brick, concrete, and tiles were measured by SEM. The results are shown in figure 2. The figures show that the surface of particles are rough, uneven and contain a lot of pores. These features indicated the surface area of the recycled particles, which is conducive to the retention and adsorption performance of the material^[3], with the use of EDS to quantitatively analyze the elements contained in the particles. The results are shown in table 1. No heavy metals were detected in the main components of the three different recycled particles.



Figure 2. The SEM images of three kinds of recycled particles Table 1. Ouantitative analysis results of three kinds of particles by EDS(w/%)

Fe
9.10

From figure 3 and 4, it can be seen that the wide range of pore size distribution of the recycled particles is a dual-model pore size distribution containing both mesopores and macropores. The nitrogen adsorption isotherm of the recycled particles is a type IV adsorption isotherm, and the relative partial pressure (P/P₀). There is an obvious H3 hysteresis ring at 0.4-0.5, which is a typical feature of mesopores. The adsorption isotherm is steeper near the saturated vapor pressure, which is caused by the multilayer adsorption in the macropores[4]. It can be seen from figure 4 that the recycled particles have a relatively concentrated distribution in the range of pore size 74.7266nm-918.4330nm, and the most probable pore size is 237.0439nm.

The results of BET analysis are shown in table 2. The specific surface areas of the BET and BJH before and after adsorption are $10.0674m_2/g$, $12.8500m_2/g$ and $8.8493m_2/g$, $9.6070m_2/g$ respectively, the specific surface area of BET and BJH decreased by 12.0994% and 25.2374%, respectively. The pore volume of the BJH was 0.0245 and 0.0229, which decrease of 6.4519%.





Figure 4. BJH pore distribution of adsorption line of recycled particles

Table 2.	Comparison	of BET	values of	recycled	particles	before an	d after	adsorption
1 4010 2.	Comparison	OI DLI	rurueb or	100,0100	pullieleb	outore un	a arter	ausorption

	Before adsorption	After adsorption
BET superficial area	10.0674	8.8493
m^2/g		
BJH superficial area	12.8500	9.6070
m^2/g		
BJH pore volume cm ³ /g	0.0245	0.0229

3.2. The kinetic adsorption characteristics

The adsorption kinetic characteristics can be described by a pseudo-first kinetic model and a pseudo-second kinetic model. The pseudo-first kinetic model is usually expressed by formula (3):

$$\frac{1}{q_t} = \left(\frac{k_1}{q_1}\right) \left(\frac{1}{t}\right) + \frac{1}{q_1}$$
(3)

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where q_1 is the equilibrium adsorption capacity (mg/g), q_t is the adsorption capacity at t (mg/g), and k_1 is the pseudo-first kinetic adsorption rate constant. pseudo-second dynamics is usually expressed by formula (4):

$$\frac{\mathbf{t}}{q_t} = \frac{1}{k_2 q_2^2} + \frac{1}{q_2} t \tag{4}$$

where q_2 is the equilibrium adsorption capacity (mg/g), q_t is the same as formula (3), and k_2 is the pseudo-second kinetic adsorption rate constant.

The adsorption of ammonia nitrogen by the recycled particles at different times showed that the adsorption amount increased rapidly at first, and the adsorption amount increased and decreased at 40-60 minutes, and then the adsorption amount increased slowly. By fitting the adsorption process according to the pseudo-first kinetics and pseudo-second formulas, it is found that the pseudo-second kinetics R^2 is greater than 0.95, indicating that the ammonia nitrogen adsorption process of the recycled particles is more in line with the pseudo-second kinetic model. The fitting curve is shown in figure 5, and the fitting parameters are shown in table 3. The fitting results of the adsorption capacity (q₂) were 0.0126mg/g, 0.0154mg/g, 0.0146mg/g, 0.0152mg/g and the actual measurement results were 0.0118mg/g, 0.0153mg/g, 0.0151mg /g is very close, and the pseudo-second kinetic can simulate the adsorption process of ammonia nitrogen by the recycled particles. This process is consistent with the process in which ions rapidly adsorb on the surface of the adsorbent and then slowly penetrate into the adsorbent, and the cations in the adsorbent gradually exchange with the ions in the solution^[5].



Figure 5. pseudo-Second kinetics adsorptionkinetic curve

Figure 6. Isothermal adsorption of ammonia by recycled particles

Table 3	. Pseudo-second k	inetic parameters of	ammonia nitrogen ad	lsorption of recycled particles
	grain size /mm	$q_2/(mg/g)$	$K_2/g(mg/min)^{-1}$	\mathbb{R}^2
	0—2	1.2618×10 ⁻²	22.4763	0.9897
	2—3	1.5357×10 ⁻²	5.6980	0.9926
	3—5	1.4583×10 ⁻²	17.1219	0.9984
	5—10	1.5245×10 ⁻²	8.5544	0.9976

3.3. Isotherm adsorption model

The Langmuir isotherm adsorption model is usually expressed as formula (5):

$$\frac{1}{q_e} = \frac{1}{C_e q_m K_L} + \frac{1}{q_m}$$
(5)

In the formula, q_e is the equilibrium adsorption capacity (mg/g), C_e is the equilibrium concentration (mg/L), q_m is the maximum adsorption capacity (mg/g), K_L is the Langmuir equation adsorption constant (L/mg), which is related to the nature of the adsorbent. The larger the K_L , the better the adsorption capacity. The value of q_m and K_L can be determined by the linear fitting of q_e and C_e . Freundlich isotherm adsorption model is usually expressed by formula (6):

$$Inq_e = InK_F + \frac{1}{n}InC_e \tag{6}$$

In the formula, q_e and C_e are the same as formula (5), and K_F is the Freundlich equation adsorption constant (mg/L), which is related to the adsorption strength.

Table 4. Parameters of isothermal adsorption model for recycled particles						
Particle size	Langmuir	Freundlich				
	$q_m (mg/g)$	$K_L (L/mg)$	\mathbb{R}^2	1/n	K _F	\mathbb{R}^2
0-2mm	0.1391	2.0584	0.9973	0.3953	0.0889	0.9787
2-3mm	0.1420	2.1452	0.9511	0.3839	0.0918	0.9315
3-5mm	0.1366	2.4870	0.9913	0.3896	0.0933	0.9901
5-10mm	0.1202	3.4577	0.9535	0.3040	0.0884	0.9082

The adsorption isotherm of the recycled particles to ammonia nitrogen is shown in figureure 6. As the equilibrium concentration increases, the equilibrium adsorption capacity first increases and then approaches saturation. The results of isotherm adsorption fitting are shown in Table 4. By observing the fitting results R^2 of the two models, it is found that the two isotherm adsorption models can describe the adsorption characteristics. The Langmuir isotherm adsorption R^2 is higher than the Freundlich, indicating that the adsorption characteristics of the recycled particles for ammonia nitrogen fits Langmuir isotherm adsorption much better than Freundlich isotherm model. Adsorption of ammonia nitrogen by the recycled particles is more inclined to the single-layer adsorption on the surface of the material. 4 particles theoretical adsorption value are 0.1391mg/g, 0.1420mg/g, 0.1200mg/g, 0.1202mg/g, which are close to the actual values of 0.1169mg/g, 0.1213mg/g, 0.1200mg/g, 0.1200mg/g, It is found after fitting that the larger the particle size, the larger the K_L value, indicating that the parameter 1/n is between 0.3040-0.3953 and less than 0.5, indicating that the adsorption of ammonia nitrogen by the recycled particle size, the recycled particles is easier^[6].

3.4. The influence of different pH on the adsorption

The results of the effect of different solution pH on the adsorption rate are shown in figure 7. It was found that the adsorption ratio of the recycled particles on the ammonia nitrogen of the different solution pH values changed slightly. When the pH value was acidic, the adsorption rate increased with the increase of the pH value. When the solution is alkaline, the adsorption rate shows a downward trend as the pH value of the solution increases.

This is because there is a dynamic conversion of $OH^++NH_4^+ \rightarrow NH_3+H_2O$ in the NH_4^+ solution with the pH change. When the solution is acidic, the solution mainly exists in the form of NH_4^+ , and the H^+ content is higher. Generally, the H^+ exchange performance is greater than NH_4^+ according to the exchange order of the ion hydration radius, which means that the more acidic the adsorbent preferentially adsorbs H^+ . When the pH of the solution becomes alkaline, there is a large amount of OH^- in the solution, and OH^- promote NH_4^+ into molecular state NH_3 with lower affinity, thereby reducing the NH_4^+ concentration and the adsorption rate. This experiment found that the maximum adsorption rate of the recycled particles for ammonia nitrogen is when the pH between 6 to 7, which is similar to the results of previous study^[7]. IOP Conf. Series: Earth and Environmental Science 647 (2021) 012160

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3.5. The influence of different ionic strength

The effect of different ionic strength on the adsorption ratio is shown in figure 8. It can be seen from the figure that as the Na⁺ increases, the adsorption rate shows a downward trend, indicating that Na⁺ in the solution compete with NH_4^+ .

 NH_4^+ and Na^+ in the solution are hydrated ions of the same valence. Generally, the adsorption of ions of the same valence by the adsorbent is related to the hydration radius and hydration energy of the ions. The closer the surface of the adsorbent, the stronger the adsorption, and the hydration radius of NH_4^+ is smaller than Na^+ , so NH_4^+ is more easily adsorbed than Na^+ [32]. When the concentration of NaCl is 0.001 mol/L, the adsorption rate of ammonia nitrogen by the recycled particles does not have a significant impact. When the concentration of NaCl reaches 0.05 mol/L, the ammonia nitrogen adsorption efficiency of the four recycled particles decreases by 19.86%, 4.95%, and 6.45 %, 8.96% respectively. When the concentration of Na^+ in the solution is twice NH_4^+ , depending on the advantage that the hydration radius of NH_4^+ is smaller than Na^+ , the recycled particles can still preferentially adsorb NH_4^+ . As the content of Na^+ continues to increase, Na^+ is predominant and the adsorption pressure is higher, which competing with NH_4^+ .

3.6.Dynamic adsorption efficiency of ammonia nitrogen in runoff by recycled particles

The dynamic adsorption efficiency of the recycled particles at different times on the ammonia nitrogen in runoff is shown in figure 9. 4 adsorption columns were sprayed at the same time, and the start time of each adsorption column was observed. The water discharge time of 0-2mm, 2-3mm, 3-5mm and 5-10mm columns were 228s, 87s, 85s, 60s respectively. It shows that the smaller the particles, the better the blocking ability of runoff, and the greater the head loss^[8]. It can be seen from figure 9 that 0-2mm, 2-3mm, 3-5mm tend to adsorb equilibrium in 60 min, and then change little. While 5-10mm tends to balance at the 30min. Contrary to static adsorption, dynamic adsorption of small and medium size recycled particles has a better adsorption effect. This is consistent with the research results of Ma Chunlin et al^[9]. [34]. It may be because the small particles have better runoff retention, which increases the adsorption rate. The large particle adsorption column has better permeability and weaker retention of runoff. Zhang Aiyuan^[10] found that the pollutant concentration of the filter column increases with the increase of the particle size of the filter material.



Figure 9. Dynamic ammonia-nitrogen adsorption efficiency of recycled particles with different particle sizes in adsorption column

3.7. Influence of particle size on adsorption rate

The static adsorption test showed that the adsorption capacity and the adsorption rate of the large recycled particles were higher than that of the small recycled particles. This may be because the adsorption of ammonia nitrogen by the recycled particles mainly depends on the capillary force and van der Waals force in the particles. Large particles can hold more solutions and ions due to the more developed pores inside. When the solid surface and internal pores contact with the solution, the additional pressure on the surface of the solution is greater than the static pressure in the direction of the pores, and the solution is gradually pressed into and fills the pores until the static pressure and the amount of additional pressure balance. In the dynamic adsorption test, due to the retardation and retention of the small particle size regeneration particles on the runoff, the liquid flow speed in the adsorption column is slowed down, and the ions have more contact with the recycled particles, so the small recycled particles have a higher adsorption capacity for ammonia nitrogen^[10].

4.Conclusion

1. The surface of the CDW recycled particles is rough and contains more mesopores and macropores. It is a safe, cheap and easily available recycled material.

2. The recycled particles have a certain adsorption capacity for ammonia nitrogen. Both the pseudo-second kinetic and Langmuir isotherm adsorption model can simulate and predict the adsorption process more accurately.

3. The adsorption rate of ammonia nitrogen increases first and then decreases at different solution pH, and the optimal adsorption rate pH is 6-7; with the increase of ionic strength, the adsorption rate of ammonia nitrogen shows a slight decreasing trend.

4. Under static adsorption conditions, large recycled particles have higher adsorption rate, while under dynamic adsorption conditions, small recycled particles have better runoff interception capacity and higher adsorption rate of ammonia nitrogen.

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