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Research Article

Removal of Ammonia Nitrogen in Aqueous Solution by the Modified Water Treatment Sludge

[1]Muqing Qiu, [2]Shuiying Xiong, [2]Haoqiang Zhou, [2]Yannan Xuan, [2]Peichao Luo,
[2]Shi Chao Ren and [2]Zebin Wang
[1]College of Life Science,
[2]Department of Life Science, Yuanpei College, Shaoxing University, Shaoxing 312000, P.R. China

Abstract: The water treatment sludge due to its high content of metal hydroxide, largely and easily available nature coupled with the fact that it is free of charge can be considered as a valuable raw material for treatment of various pollutants in wastewater. In this research, the potential and effectiveness of the water treatment sludge was studied as an alternative adsorbents for the removal of ammonia nitrogen in aqueous solution. The water treatment sludge was activated by KOH. Then, the adsorption isotherms and adsorption kinetics of the model compound over the modified water treatment sludge were determined and discussed in detail. The results showed that the equilibrium data fitted with the Langmuir isotherm. The adsorption data fitted the pseudo-second-order model.

Keywords: Adsorption isotherm, adsorption kinetic, ammonia nitrogen, water treatment sludge

INTRODUCTION

In recent years, with the expansion of industrial scale and intensification of human activities, emissions of domestic sewage and industrial wastewater containing ammonia nitrogen have been increased rapidly (Zhang *et al*., 2013). The wastewater containing ammonia nitrogen may cause eutrophication and produce toxic substances if they were discharged into the aquatic ecosystem (Camargo and Alonso, 2006). Therefore, it is imperative to remove ammonia nitrogen in aqueous solution. The research on the removal of ammonia nitrogen in aqueous solution has been carried out extensively in recent years. The main methods of removing nitrogen in aqueous solution are precipitation, crystallization, biological removal, adsorption and ion exchange (Huo *et al*., 2012; Mena-Duran *et al*., 2007; Zhao *et al*., 2006; Yang *et al*., 2011). The adsorption technology is a more effective way to remove nitrogen, due to its advantages, namely small floor area, simple process and high efficiency, among others. Therefore, it is required to develop new material for effective removal of ammonia nitrogen with low cost.

Water treatment sludge is generated from water treatment plants during the water treatment process of chemical coagulation, flocculation, sedimentation and rapid sand filtering (Huang and Wang, 2013). The disposal of water treatment sludge requires careful consideration if it is to be managed in an environmentally acceptable and sustainable manner. The water treatment sludge due to its high content of metal hydroxide, largely and easily available nature coupled with the fact that it is free of charge can be considered as a valuable raw material for treatment of various pollutants in wastewater (Vinitnantharat *et al*., 2010; Moghaddam *et al*., 2010).

In this research, the potential and effectiveness of the water treatment sludge was studied as an alternative adsorbents for the removal of ammonia nitrogen in aqueous solution. The water treatment sludge was activated by KOH. Then, the adsorption isotherms and adsorption kinetics of the model compound over the modified water treatment sludge were determined and discussed in detail.

MATERIALS AND METHODS

Preparation of the adsorbents: The water treatment sludge was obtained from water treatment plant of Shaoxing in Zhejiang Province of P.R. China. The water treatment sludge was dried at 378 K for 12 h in order to achieve constant weight. Then they were grounded and sieved into a uniform size of 100 meshes. The 50 g of the water treatment sludge was soaked stilly with 100 mL 5% KOH solution in 250 mL Erlenmeyer flasks for 12 h at room temperature. Then, it was dried again at 378 K for 12 h to constant weight and was carbonized at 873 K in a muffle furnace for 12 min. The product of 100 mesh modified water treatment sludge was thus obtained and then stored for later adsorption experiments.

Corresponding Author: Shuiying Xiong, Department of Life Science, Yuanpei College, Shaoxing University, Shaoxing 312000, P.R. China, Tel./Fax: 86-575-88345098

Adsorption experiments: Adsorption experiments were conducted in a set of 250 mL Erlenmeyer flasks containing 1.0 g of modified water treatment sludge and 100 mL of ammonia nitrogen with various initial concentrations (20, 50, 100, 150 and 200 mg/L, respectively) in aqueous solution. The flasks were placed in a shaker at a constant temperature of 308 K and 200 rpm. The samples were analyzed by the Nessler reagent spectrophotometric method.

Analytical methods: The textural characteristics of modified water treatment sludge including surface area, pore volume, pore size distribution were determined using standard N_2-adsorption techniques.

The surface physical morphology of modified water treatment sludge was observed by a scanning electron microscope. The amount of adsorbed ammonia nitrogen q_t (mg/g) at different time, was calculated as follows:

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

where C_0 and C_t (mg/L) are the initial and equilibrium liquid-phase concentrations of ammonia nitrogen, respectively. V (L) is the solution volume and m (g) is the mass of adsorbent used.

Statistical analyses of data: All experiments were repeated in duplicate and the data of results were the mean and the Standard Deviation (S.D.). The value of the S.D. was calculated by Excel Software. All error estimates given in the text and error bars in figures are standard deviation of means (mean±S.D.). All statistical significance was noted at $\alpha = 0.05$ unless otherwise noted.

RESULTS AND DISCUSSION

Characterization of modified water treatment sludge: The surface of modified water treatment sludge was observed using scanning electron microscopy as shown in Fig. 1. It can be seen from the micrograph that the modified water treatment sludge contains porous structures and high surface area. The characteristics of modified water treatment sludge are obtained from the standard N_2-adsorption techniques. The BET surface area is 124 m^2/g, the total pore volume is 0.48 cm^3/g and the Nominal pore size is 0.52 nm.

Effect of contact time and adsorption kinetics: The influence of contact time on the removal of ammonia nitrogen in aqueous solution by the modified water treatment sludge is shown in Fig. 2. The adsorption rate of ammonia nitrogen increases sharply at short contact time and slowed gradually as equilibrium was approached. From Fig. 2, it can be also concluded that the equilibrium time is 360 min. It may be due to the availability of initial large number of vacant surface

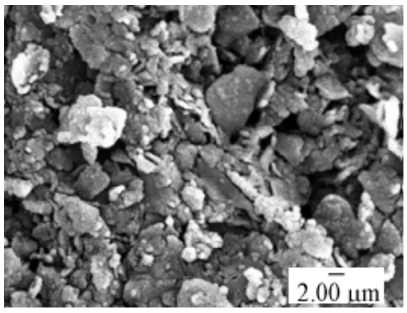

Fig. 1: SEM image of modified water treatment sludge

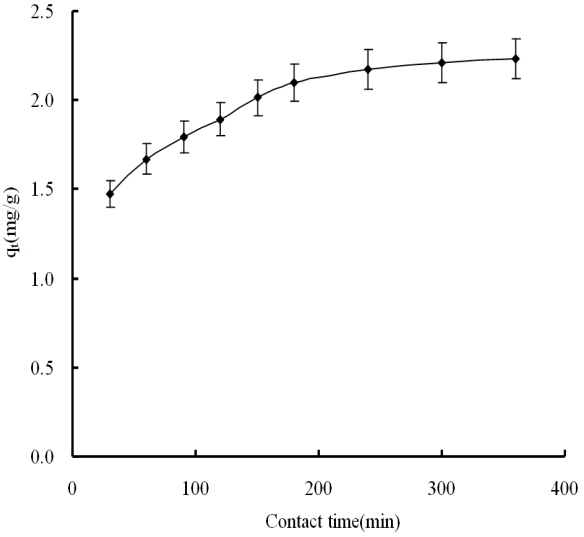

Fig. 2: Effect of contact time on removal of ammonia nitrogen ions
Experiment concentration: 1.0 g of the modified water treatment sludge, 50 mg/L ammonia nitrogen ions, 308 K and 200 rpm

active sites for adsorption and adsorption rate is very fast. As equilibrium was approached, the filling of vacant sites becomes difficult due to repulsive forces between ammonia nitrogen adsorbed on solid surface and ammonia nitrogen from solution (Thinakaran *et al*., 2008).

The linear pseudo-first-order kinetic model of Lagergren is given as follows (Thinakaran *et al*., 2008):

$$\ln(q_e - q_t) = \ln q_e - k_1 \times t \quad (2)$$

where, q_e and q_t are the amounts of ammonia nitrogen ions absorbed onto the adsorbent (mg/g) at equilibrium and at t respectively. k_1 is the rate constant of first-order adsorption (min^{-1}).

The pseudo-second-order kinetic model developed by Ho and McKay (1998) is based on the experimental information of solid-phase sorption. The linear pseudo-second-order model can be expressed as follows:

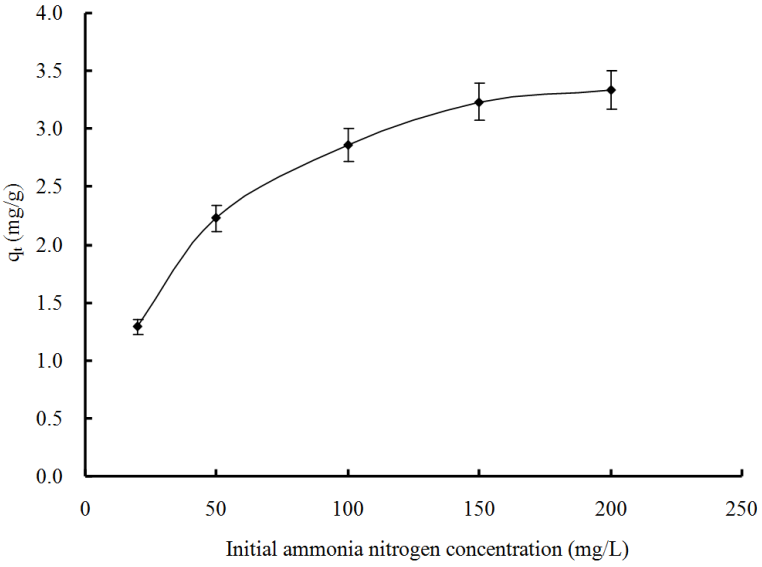

Fig. 3: Effect of initial ammonia nitrogen concentration on adsorption capacity
Experiment concentration: 1.0 g of modified water treatment sludge, contact time of 360 min, 308 K and 200 rpm

Table 1: The kinetic parameters for the adsorption of ammonia nitrogen ion on the modified water treatment sludge

Pseudo-first-order			Pseudo-second-order		
q_e (mg/g)	k_1 (min^{-1})	R^2	q_e (mg/g)	k_2 (g/mg/min)	R^2
1.17	0.003	0.9169	2.39	0.016	0.9989

Experimental conditions: 1.0 g of modified water treatment sludge, 50 mg/L ammonia nitrogen ions, 308 K and 200 rpm

Table 2: The adsorption isotherm parameters for the adsorption of ammonia nitrogen ion on the modified water treatment sludge.

Langmuir			Freundlich		
q_m (mg/g)	K_L (L/mg)	R^2	K_f (mg/g)	n	R^2
3.49	0.28	0.9985	2.67	0.27	0.9483

Experimental conditions: 1.0 g of modified water treatment sludge, contact time of 360 min, 308 K and 200 rpm

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (3)$$

where, k_2 is the rate constant of second-order adsorption (g/mg/min).

According to Eq. (2) and (3), the kinetic parameters of pseudo-first-order kinetic model and pseudo-second-order kinetic model for adsorption of ammonia nitrogen ion on the modified water treatment sludge were calculated. The experiment data came from Fig. 2. Table 1 is the kinetic parameters for adsorption of ammonia nitrogen ion on the modified water treatment sludge. The adsorption data fitted the pseudo-second-order model much better with R^2 greater than the pseudo-first-order model. It implies that the predominant process is chemisorption, which involves a sharing of electrons between the adsorbate and the surface of the adsorbent.

Effect of initial ammonia nitrogen concentration and adsorption isotherm: The effect of initial ammonia nitrogen concentration on the ammonia nitrogen removal efficiency was investigated. The tests of results were shown in Fig. 3. As shown in Fig. 3, the adsorption capacity of ammonia nitrogen on modified water treatment sludge increased with increasing initial ammonia nitrogen concentration in solution.

The adsorption isotherm was described with Langmuir isotherm and Freundlich isotherm. The Langmuir isotherm equation is represented by the following Eq. (4):

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad (4)$$

where,

C_e = The equilibrium concentration of ammonia nitrogen ions (mg/L)
q_e = The amount of ammonia nitrogen ions adsorbed (mg/g)
q_m = The maximum adsorption capacity of ammonia nitrogen ions (mg/g)
K_L = The Langmuir adsorption equilibrium constant (L/mg) related to the affinity of the binding sites (Langmuir, 1918)

The Freundlich isotherm equation is described by the following Eq. (5):

$$q_e = K_F C_e^{\frac{1}{n}} \quad (5)$$

where, K_F and n are the Freundlich adsorption isotherm constants, which are indicators of adsorption capacity and adsorption intensity respectively (Freundlich, 1906).

Langmuir and Freundlich isotherms were fitted to the experimental data from Fig. 3. The corresponding constants were calculated according to Eq. (4) and (5), which were listed in Table 2. The results indicated that the Langmuir isotherm fitted better than the Freundlich isotherm for the adsorption of ammonia nitrogen ion on the modified water treatment sludge. The adsorption process is heterogeneity of the adsorbents and favorable adsorption.

CONCLUSION

The potential and effectiveness of the water treatment sludge was studied as an alternative adsorbents for the removal of ammonia nitrogen in aqueous solution. The water treatment sludge was activated by KOH. The modified water treatment sludge contains porous structures and high surface area. The BET surface area is 124 m^2/g, the total pore volume is 0.48 cm^3/g and the Nominal pore size is 0.52 nm. The adsorption data fitted the pseudo-second-order model much better with R^2 greater than the pseudo-first-order model. It implies that the predominant process is chemisorption, which involves a sharing of electrons between the adsorbate and the surface of the adsorbent. The Langmuir isotherm fitted better than the Freundlich isotherm for the adsorption of ammonia nitrogen ion on the modified water treatment sludge. The adsorption process is heterogeneity of the adsorbents and favorable adsorption.

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