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Material prepared from drinking waterworks sludge as adsorbent for ammonium removal from wastewater

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ABSTRACT

Drinking waterworks sludge (DWS) is not an effective adsorbent for ammonium removal without any treatment. In this study, DWS was used as a starting material to prepare ammonium adsorbent (M-DWS) by means of an ultrasonic assisted extraction and synthesis method. Two materials (M-DWS1# and M-DWS2#) were prepared according to two different routes. The composition, structure, and surface properties of DWS and M-DWS were characterized and their ammonium adsorption abilities were examined. Characterization results showed that the lamellar structure of DWS was converted into the spherical units of M-DWS and that the cation exchange capacity and specific surface area of M-DWS were many times higher than that of DWS. Batch test results indicated that the adsorption equilibrium data of M-DWS fitted well to both the Langmuir and Frendlich isotherms. The maximum adsorption capacity of M-DWS1# and M-DWS2# evaluated from the Langmuir isotherm was 6.11 mg/g and 5.10 mg/g, respectively. It was also observed that the initial pH affected ammonium adsorption on M-DWS greatly. Under an optimum pH of 7–8, the highest ammonium removal rate of 90% for M-DWS1# and 80% for M-DWS2# were achieved at an initial concentration of 50 mg NH_4^+/L. The advantage of M-DWS2# lies in its higher yield and less waste discharge compared with M-DWS1#.

1. Introduction

Drinking water works sludge (DWS) is an inevitable water industrial waste. Large quantities of DWS are produced everyday in China, as among most other countries worldwide. DWS has been disposed mainly by landfill, incineration and ocean discharge. In recent decades, the reuse of DWS has catched scholar's interest and attention as sludge disposal cost increases and landfill capacity decreases [1,2]. DWS has been studied extensively as a low-cost adsorbent for removing some pollutants such as phosphorus [3,4], arsenic [5], boron [6], and fluoride [7] from wastewater based on its main components including silicon, aluminum, and iron oxides as well as some clay minerals. It has also been indicated that specific adsorption may be responsible for the removal of these anionic contaminants by DWS and that ligand exchange should be the main adsorption mechanism [8,9]. DWS, however, does not exhibit affinity for cationic pollutants such as ammonium because of a different adsorption mechanism.

Ammonia nitrogen is also a major pollutant in water. It is stated in Environmental Quality Standards for Surface Water in China that the ammonia nitrogen concentration for first and second class surface water sources should not exceed 0.15 and 0.50 mg/L, respectively. However, the ammonia nitrogen concentration of some surface water serving as a source of potable water is much higher than the limits value of standard due to large quantities of industrial and municipal wastewater discharged into existing water resources. In view of the adverse environment effect, the discharge standards of ammonia nitrogen have also been established for municipal wastewater treatment plants in China and it was set to 5 and 8 mg N/L for A and B standard of first grade effluent, respectively, and 25 mg N/L for second grade effluent. Therefore, removal of ammonium from surface water and wastewater has been given great consideration [10]. Among various methods available for ammonium removal, ion exchange adsorption has received great attention from scientists with the development of high-efficient ion exchangers such as zeolite [11]. Moreover, several works related to low-cost ammonium adsorbents prepared from natural mineral [12] and industrial wastes [13] by means of a conventional hydrothermal method has been cited in literatures. Nevertheless, to the best of our knowledge, no previous effort has been made to

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prepare ammonium adsorbent using DWS as a starting material. We suggest that the potential of DWS to be an effective adsorbent for ammonium removal should be assessed, partly due to its similarities in chemical compositions with those materials used in preparing ammonium adsorbent such as fly ash and clay, but even more so due to its most recognized feature of being local, easy, and largely available. Therefore, the aim of this study was to prepare effective ammonium adsorbent using DWS. This will not only promote the beneficial reuse of DWS in wide range of wastewater treatment but also provide an alternative material for ammonium removal.

Based on the above analysis, DWS may be not directly useful for ammonium removal and it is likely due to its lower cation exchange capability (CEC). This should be associated with the surface characteristics of DWS such as surface type, surface charge and surface area. Thus, the treatment for DWS was focused on the improvement of CEC through changing its surface properties. A simple and efficient approach learned from a conventional hydrothermal method was proposed. It was mainly composed of extraction and synthesis process with the assistance of ultrasonic. Ultrasound has proven to be a powerful tool for enhancing chemical processes such as solid–liquid extraction [14] and material synthesis [15,16]. In our experiment, an acid and alkali treatment was performed in an ultrasonic extraction apparatus for enhancing the extraction efficiency of aluminum and silicon from DWS [4]. Furthermore, an ultrasonic generator was used to accelerate the process of ammonium adsorbent preparation [16]. Finally, two materials were prepared according to two slightly different routes for making appropriate choices in their practical application based on the comprehensive consideration of material performance, the ratio of raw material utilization and the amount of waste discharged.

The objectives of this study were as follows: (1) to prepare ammonium adsorbents (M-DWS) using DWS by means of the proposed method; (2) to characterize the structure and surface properties of M-DWS; (3) to examine the ammonium adsorption capacity of M-DWS; (4) to analyze the reasons for the improved performance of M-DWS.

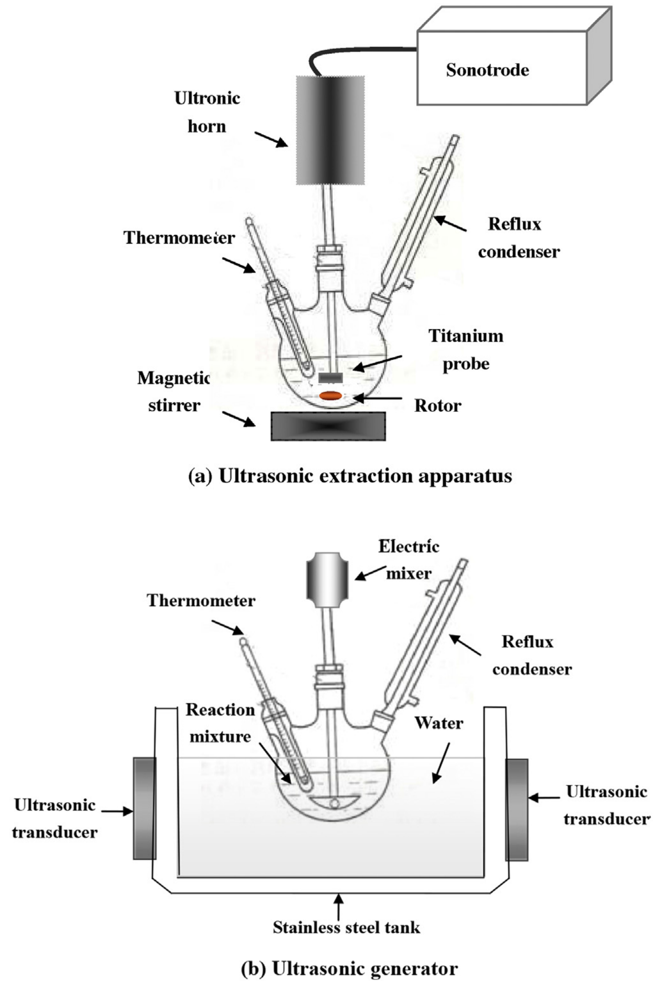

Fig. 1. Experimental set-up for preparation of adsorbent using (a) ultrasonic extraction apparatus and (b) ultrasonic generator.

2. Materials and methods

2.1. Preparation of sludge samples and synthetic wastewater

The raw DWS used in our test was supplied by the New District Waterworks of Suzhou, China. This plant utilizes both iron and aluminum salts as coagulants to flocculate suspended particles and colloids during drinking water treatment. DWS was allowed to air-dry at room temperature (25 °C) for about 4 weeks in our laboratory. The dried DWS was ground in a grinder and subsequently homogenized by sieving. The DWS sample was prepared as a starting material for the preparation of ammonium adsorbent.

The experimental set-up with ultrasonic extraction apparatus and ultrasonic generator is depicted in Fig. 1. The flow chart describing the preparation process is shown in Fig. 2. There were two slightly different routes adopted for the preparation of ammonium adsorbent from DWS. In the route 1, DWS was treated in a sulfuric acid solution of 2 M at a solid/liquid ratio of 1:20 using an ultrasonic extraction apparatus, which generated ultrasonic through a titanium probe at a frequency of 20 kHz and a power of 1000 W, with continuous stirring for 0.5 h. After acid treatment, the suspension was centrifuged and the sediment I was mixed with a sodium hydroxide solution of 20 M at a solid/liquid ratio of 1:1.5 in an ultrasonic extraction apparatus working at 600 W and 20 kHz, with continuous stirring for 1 h. After alkali treatment, the suspension was centrifuged and in a round flask the supernatant II was mixed with a certain amount of the supernatant I, by which the pH of the resulting mixture was controlled at 7–8. Then the flask was immersed in an ultrasonic generator, which consists of stainless-steel tank filled with water and two ultrasonic transducers attached to the external wall of the tank. Ultrasonic frequency and power of the tank fixed at 40 kHz and 400 W, and ultrasonic energy was transmitted to the material inside the flask through water (65–75 °C). After treating 4 h, the product was isolated with filtration, washed with water until pH reached 7, and dried at 105 °C.

In the route 2, the acid and alkali treatment were the same as performed in the route 1. Different to the route 1, the sediment I after alkali treatment was mixed directly with a certain amount of the supernatant I without centrifugation. The subsequent treatment followed the same procedures as in the route 1. The samples obtained by the route 1 and route 2 were called M-DWS1# and M-DWS2#, respectively. It can be found from Fig. 2 that the sediment II was discarded as waste in the route 1 and that there was little waste in the preparing of M-DWS2#.

The synthetic wastewater containing ammonium was prepared using anhydrous NH_4Cl. The ammonium concentration in municipal wastewater is typically about 50 mg/L. Accordingly, the concentration of the synthetic ammonium wastewater used in our test ranged from 10 to 90 mg/L, which is similar to that of the actual wastewater treated in the FuXing Wastewater Treatment Plant of Suzhou, China.