

Phosphate adsorption performance of a novel filter substrate made from drinking water treatment residuals

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ABSTRACT

Phosphate is one of the most predominant pollutants in natural waters. Laboratory experiments were conducted to investigate the phosphate adsorption performance of a (NFS) made from drinking water treatment residuals. The adsorption of phosphate on the NFS fitted well with the Freundlich isotherm and pseudo second-order kinetic models. At pH 7.0, the maximum adsorption capacity of 1.03 mg/g was achieved at 15°C corresponding to the wastewater temperature in cold months, and increased notably to 1.31 mg/g at 35°C. Under both acidic conditions (part of the adsorption sites was consumed) and basic conditions (negative charges formed on the surface of NFS, which led to a static repulsion of PO_4^{3-} and HPO_4^{2-}), the adsorption of phosphate was slightly inhibited. Further study showed that part of the adsorption sites could be recovered by 0.25 mol/L NaOH. The activation energy was calculated to be above 8.0 kJ/mol, indicating that the adsorption of phosphate on NFS was probably a chemical process. Considering the strong phosphate adsorption capacity and recoverability, NFS showed great promise on enhancing phosphate removal from the secondary treated wastewater in the filtration process.

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Introduction

Phosphate is one of the critical pollutants in natural waters (Fulazzaky et al., 2014). It is mainly produced as the result of human activities, and ultimately enters water bodies. The existence of phosphate will change the trophic state of natural waters and lead to algal bloom (Muscutt and Withers, 1996; Yang et al., 2013), which adversely affect the water quality in terms of esthetic problems (Ragheb, 2013), changing oxygen content, and thus, leading to the death of aquatic livings (Biswas et al., 2007). Besides, toxins are

produced as the growth of algae. These toxins can transfer into the human body through the food chain (Yuan et al., 2006). Thus, the content of phosphate in the water environment must be controlled to maintain the ecological balance of the natural system (Conley et al., 2009).

To enhance the removal of phosphate from the wastewater, lots of methods have been developed (Hui et al., 2014; Vaiopoulou et al., 2007). Biological treatment is the most commonly used method and shows an advantage of low cost. However, the removal rate of phosphate is not stable (Vaiopoulou et al., 2007). Accordingly, chemical treatment

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methods such as the addition of calcium or magnesium salts are used to guarantee phosphate removal. However, the costs of the chemical reagents are usually high, and the treatment and disposal of the chemical sludge should be considered. Besides chemical precipitation, adsorption is used to remove phosphate because of its high efficiency and easy operation (Bhattacharyya and Gupta, 2008; Palanisamy and Sivakumar, 2009; Wajima and Rakovan, 2013). Materials such as activated carbon (Zhang et al., 2011), natural clays (Mangwandi et al., 2014; Wei et al., 2014), modified clays (Yang et al., 2014; Zamparas et al., 2012), and industrial wastes (Barca et al., 2012; Rhue and Harris, 1999) have been used for phosphate removal to guarantee an advanced treated water quality.

Drinking water treatment residuals (WTR) are produced globally as a by-product of the coagulation process. Major components of WTR are soils, organic materials, and metal hydroxides/oxides, depending on the metal salt used for coagulation (Babatunde and Zhao, 2007). For the residuals produced in the plants using aluminum-based coagulants, Al is the major metal component with a weight ratio of $12\% \pm 2\%$ (Ippolito et al., 2011). Other elements, such as Fe, Ca, Mg, Cr, Cu, Cd, Ni, Pb, and Zn were also detected but with a much lower weight ratio. These wastes are usually disposed off to landfills (Makris and O'Connor, 2007; Nair and Ahammed, 2015). Recently, efforts have been made to reuse WTR in water and wastewater treatments (Guan et al., 2005). Gibbons et al. (2009) studied the reuse of WTR as a coagulant for phosphate removal. It was proved to be a cost-effective adsorption material for treating wastewaters with high phosphate contents (Babatunde et al., 2011; Gao et al., 2013). However, the use of WTR as a potential filter substrate to achieve an enhanced phosphate removal has not yet been reported.

In this study, WTR were used as the major material in the production of a novel filter substrate (NFS). The aim of this work is to investigate the phosphate adsorption performances of the NFS and the possibility of utilizing the NFS as a filter substrate for the enhanced removal of phosphate from secondary treated wastewaters.

1. Experimental materials and methods

1.1. Preparation of filter substrate

The NFS used in this study was prepared from the WTR produced by a local plant in Xi'an, China. The plant is fed by the Heihe Reservoir with a total water supply capacity of 450,000 m³/day. The average pH, turbidity, and total organic carbon (TOC) in the raw water was 7.6, 23.8 NTU, and 5.4 mg/L, respectively. Poly-aluminum chloride (PACl) is used as the coagulant, and the flocs formed in coagulation were the major material of NFS. Besides WTR, kaolin and humic acid were also introduced. Kaolin after calcining shows a great pozzolanic property that can enhance the strength of materials (Sabir et al., 2001). Humic acid, with a linear aggregation structure under basic conditions (He et al., 2008), will be oxidized into CO2 and H₂O after calcining and produce quantities of micro-pore structures, which helps to improve the specific surface area of NFS. Based on the target density (2.1×10^3 – 2.3×10^3 kg/m³) and specific area of the NFS, the weight ratio of WTR:kaolin:humic acid was optimized. The preparation process was as follows: (1) drying the sludge to a constant weight at 105°C, (2) mixing the sludge trituration with kaolin and humic acid (Aldrich Chemical Company, USA) in a weight ratio of 10:7:2, (3) adding certain amount of deionized water to maintain a water ratio of 60%-65%, (4) NFS with equivalent diameters of 2–4 mm were made by heating the sludge balls at 200°C for 1.0 hr, and then 650°C for 2.0 hr.

1.2. Effects of pH and temperature on the adsorption of phosphate

Adsorption tests were conducted to investigate the effects of pH (at 25°C) and temperature (at pH 7.0) on the removal of phosphate, respectively. Solutions of 2.0 mg/L phosphate (50 mL) were added to test bottles with 0.5 g of NFS. HCl and NaOH solutions (0.2 mol/L) were used to adjust the pH of the adsorption system. The concentration of phosphate residual in the solution was determined after 24 hr of adsorption. Meanwhile, the speciation and solubility of phosphate in solutions with Al(III) co-existing were calculated using the VISUAL MINTEQ software (KTH Royal Institute of Technology, Sweden). All the input parameters including suspension pH, water temperature, NFS dosage, and salt contents were the same as that controlled in the adsorption tests.

1.3. Adsorption isotherm experiments

NFS weighing 0.5 g was added into 50 mL test bottles. Phosphate solutions were prepared with Na_3PO_4 and then transferred into the test bottles. The adsorption isotherm was measured with initial phosphate concentrations ranging from 1.0 to 10.0 mg/L at pH 7.0. The test bottles were placed in water bath for 24 hr at a constant temperature. Blank solution without NFS was also included. After achieving equilibrium, 10 mL water samples were taken and pretreated with a 0.45 μ m filtration membrane prior to analysis. All of the experiments were conducted three times. The adsorption capacity of NFS for phosphate q_e (mg/g) was calculated as described in Eq. (1):

$$q_e = \frac{(C_i - C_e) \times V}{m} \tag{1}$$

where, C_i (mg/L) and C_e (mg/L) are the concentrations of phosphate at initial time and when the adsorption system reached equilibrium, respectively; V (mL) is the total volume of the test solution, *m* (g) is the mass of the NFS added to the test solution, respectively. Langmuir and Freundlich models, as described in Eqs. (2) and (3) respectively (Xiong et al., 2008), were used to describe the adsorption performances of NFS for phosphate.

$$\frac{C_{\rm e}}{q_{\rm e}} = \frac{1}{k_{\rm a}q_{\rm m}} + \frac{C_{\rm e}}{q_{\rm m}} \tag{2}$$

$$\log q_{\rm e} = \log K_{\rm f} + \frac{1}{n} \log C_{\rm e} \tag{3}$$

where, q_m (mg/g) is the maximum adsorption capacity of the NFS; k_{a_r} (L/mg) is adsorption constant, K_f (mg/L) is the Freundlich constant, and *n* (dimensionless) is the Freundlich exponent.

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