



Adsorptive removal of phosphorus from aqueous solution using sponge iron and zeolite

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ABSTRACT

Phosphorus adsorptive removal is an important and efficient treatment process in constructed subsurface flow wetlands. Many materials have been proposed for removal of excess phosphorus from wastewater. Selecting a substrate with a high phosphorus adsorption capacity is therefore important in obtaining significant phosphorus removal. In this study, the phosphorus removal capacities of sponge iron and zeolite were evaluated and related to their physico-chemical characteristics. The potential mechanisms affecting the adsorptive removal of phosphorus from aqueous solutions onto sponge iron and zeolite were investigated in batch experiments. The pseudo-second-order kinetics were useful since the adsorption rate data fitted well. The Freundlich and Langmuir models well described the adsorption isotherm data. The results of static experiments and dynamic experiments (column experiments) indicated that the adsorption of phosphorus onto sponge iron was more apt to chemical combination, but zeolite was more apt to electrostatic attraction or ion-exchange. For sponge iron, some iron (iii) (Fe^{3+}) or iron (ii) (Fe^{2+}) and phosphate ions (P) form Fe-P, the solid phases compound was fixed. For zeolite, aluminum oxide and silicon oxide formed complexes in aqueous solution. It was observed that positive or negative charge surface sites favored the adsorption of phosphate due to the electrostatic attraction or ion-exchange.

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1. Introduction

Constructed wetlands have been described as “environmentally sensitive and cost-effective treatment systems for wastewater renovation” [1]. They can be thought of as “ecological engineers” [2] because the concept involves supplying the necessary components and capitalizing on the naturally occurring wetland processes to reduce the targeted pollutants. Constructed wetlands are widely used to treat a wide variety of wastewater throughout the world because of their low cost, low energy consumption, and good effect they promise in wastewater treatment [3]. Using constructed wetlands for the treatment of polluted water is increasing in popularity as an ecological engineering alternative to conventional and chemical based methods.

The substrate, vegetation, and microbial communities all make up a wetland ecosystem. Substrates capture high levels of total phosphorus from the effluent. The concentration of total phosphorus is thus reduced after adsorption process. The choice of substrates is vital in wastewater treatment by means of constructed wetlands [4]. In phosphorus removal, the appropriate choice of filling substrate is a key factor [5]. It is important to select substrates with a high phosphorus immobility capacity and economical

friendliness in order to obtain a sustainable phosphorus removal in constructed wetlands.

Recently, the removal of phosphorus from aqueous solutions and wastewater via substrates has been widely studied [6–13]. It is generally found that several of these substrates have the potential to enhance the phosphorus removal in constructed wetland systems. Our studies aimed at selecting substrates with a high phosphorus sorption capacity. Based on the static and dynamic experiments, this paper analyzes the adsorption capacity of two kinds of substrates: sponge iron and zeolite. Zeolite is an excellent and traditional adsorbent, but its application in phosphorus removal in wastewater treatment is barely reported. Sponge iron is also a traditional environmental material, but its application as an adsorbent in phosphorus removal is a new research field.

During the study, the adsorption capacity and practical applicability of phosphorus adsorption process were explored based on the experimental data. The Langmuir and Freundlich isotherm models were used to fit the experimental data and tested for their applicability. The pseudo-second-order kinetic models were employed to interpret the kinetics results under static conditions. The practical applicability of the sponge iron and zeolite in wastewater treatment was analyzed experimentally under dynamic conditions. This paper explains the science behind sponge iron and zeolite, and their immediate and long-term advantages over conventional substrates.

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2. Materials and methods

2.1. Materials

Sponge iron and zeolite were obtained from Henan Gongyi Huanglong Filter Factory, China. Sponge iron was composed of 95% Fe₂O₃ and other oxides; the specific surface area was ≥80 m²/g. While zeolite was composed of 5% Al₂O₃ and 95% SiO₂, the specific surface area was 500–800 m²/g. The two adsorption substrates were washed several times with distilled water to remove any surface-adhered particles and to be sure that there were no soluble salts which could be dissolved during the batch studies. The washed substrates were then dried to a constant weight in an oven at 105 °C for 48 h. They were later converted into fine powder by grinding with a mechanical grinder. They were sieved with a 20-mesh (0.85 mm) particle size sieve. Finally, they were stored in a desiccator for further use [14].

All reagents were of analytical grade, obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). In addition, all solutions were prepared using deionized water. Stock solutions of phosphorus (1000 mg L⁻¹) were prepared using anhydrous potassium dihydrogen phosphate (KH₂PO₄). The pH was pre-adjusted by HCl and NaOH solutions and measured by a pH meter with a combined pH electrode. The stock solution was diluted appropriately as necessary. The concentrations of the phosphorus were determined using the ascorbic acid method with a Unico Spectrophotometer (US-2102 PCS) [15]. To analyze phosphorus removal capacity and practical applicability of sponge iron and zeolite in wastewater treatment, samples were collected from Guangdong Heyuan Sewage Treatment Plant. In the wastewater, the initial concentrations of TP, TDP, PP, SRP, and DOP were 2.0–2.5 mg/L, 1.5–2.0 mg/L, 0.2–0.5 mg/L, 1.5–2.0 mg/L, and 0.1–0.5 mg/L respectively.

2.2. Phosphorus analysis

Total phosphorus (TP) in water can be separated into particulate phosphorus (PP) and total dissolved phosphorus (TDP). TDP can further be separated into inorganic and organic phosphorus. Whereas soluble reactive phosphorus (SRP) is an inorganic type, dissolved organic phosphorus (DOP) is organic in nature [16]. These different forms of phosphorus were determined and analyzed. TP was determined after 30 min of autoclave-mediated digestion (120 °C, 100 kPa, with K₂S₂O₈ and H₂SO₄) of an unfiltered sample. The molybdenum-blue method was employed in photometric TP analysis. A separate analysis of TDP (a sample of 0.45-μm filtrate digested and determined as in the TP analysis) that would also determine PP by subtracting the TDP from TP was employed [17]. SRP was analyzed using the molybdenum-blue method [18]. The sample was mixed with a reagent containing ammonium molybdate, potassium tartrate, and sulfuric acid. Ascorbic acid was then added, and a blue complex formed was measured using a flow through detector. For the determination of TDP, organic phosphorus was first oxidized to SRP with the addition of an alkaline potassium persulfate reagent. This was coupled with heating and UV oxidation. Polyphosphates were oxidized to SRP by sulfuric acid digestion. The sample was then analyzed in a similar manner to SRP. DOP was calculated by subtracting SRP from TDP [19]. All the above procedures were repeated, and the average values were used for analysis.

2.3. Static experiments

The phosphorus static experiments, including the kinetic studies, adsorption isotherms and desorption experiment, were carried out by batch experiments in 100 mL conical flasks. The flasks were

capped and shaken horizontally on a shaker equipped with thermostat at 120 rpm at 25 ± 1 °C. This was followed by the filtration of the aqueous supernatants through a 0.45 μm membrane filter. The concentrations of phosphorus in the filtrate samples were spectrophotometrically determined. All the above procedures were repeated for several batches, and the average values used for analysis.

2.3.1. Adsorption isotherms

Isotherm data were analyzed using Langmuir and Freundlich adsorption equations. The Langmuir and Freundlich parameters were determined and correlation coefficients calculated. The linear form of the Langmuir isotherm is represented below [20]:

$$\frac{C_e}{Q_e} = \frac{1}{Q_m k_1} + \frac{C_e}{Q_m} \quad (1)$$

where Q_e is the equilibrium phosphorus concentration on adsorbent (mg/kg), C_e is the equilibrium phosphorus concentration in solution (mg/L), Q_m is the monolayer phosphorus adsorption capacity of the adsorbent (mg/kg), and k_1 is binding constant (L/mg) [21].

The linear form of Freundlich isotherm model can be written as [20]:

$$\lg Q_e = \lg K_F + \frac{1}{n} \lg C_e \quad (2)$$

where K_F and $1/n$ are the Freundlich constants which are related to the adsorption capacity ((mg/kg)/(mg/L)^{1/n}) and adsorption intensity respectively. Q_e is still the equilibrium phosphorus concentration on adsorbent (mg/kg).

Adsorption isotherm study was carried out by shaking 5 g of substrates (20 mesh) in 50 mL KH₂PO₄ solutions with different initial concentrations (3, 6, 10, 12, 15, 18, 24, and 40 mg/L) for 48 h.

2.3.2. Adsorption kinetics

The pseudo-second-order kinetic expression for adsorption systems has been applied in a number of studies including some involving adsorption of anions [22]. Kinetics of the phosphorus ions adsorption was described using a pseudo-second-order mechanism. The linear form of the model is given as follows [23]:

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

where k_2 is the rate constant of pseudo-second-order kinetic model (kg/mg h). The values of equilibrium adsorption capacity (Q_e) and rate constant (k_2) are calculated from the intercept and the slope of the linear plot of t/Q_t versus t , along with the value of determination coefficient, R^2 .

Adsorption experiments for the kinetic study was carried out by shaking 5 g of substrates (20 mesh) in 50 mL KH₂PO₄ solutions with an initial concentration of 12 mg/L. The set experimental contact times were 2, 4, 8, 12, 18, 24, 30, and 48 h.

2.3.3. Phosphorus desorption

Desorption experiments conducted to explore the reversibility of the sorption reactions were carried out with pre-sorbed P saturated substrates. Pre-sorbed P samples were prepared by the phosphorus adsorption experiments. To maximize the amount of phosphorus that the sponge iron and zeolite would remove from aqueous solution under our study conditions, the phosphorus desorption kinetics in the presence of the substrates was determined. Based on phosphorus desorption experiment, the properties of adsorption such as physical, chemical or both, would be investigated.

The sponge iron and zeolite desorption process was also studied. The rate of phosphorus desorption from the substrates was

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