Journal of the Taiwan Institute of Chemical Engineers 000 (2017) 1–6

Contents lists available at ScienceDirect

Journal of the Taiwan Institute of Chemical Engineers

journal homepage: www.elsevier.com/locate/jtice

Removal performance of phosphate from aqueous solution using a high-capacity sewage sludge-based adsorbent

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ARTICLE INFO

Article history:
Received 25 October 2016
Revised 2 March 2017
Accepted 5 April 2017
Available online xxx

Keywords:
Adsorption
Sewage sludge-based adsorbent
Phosphate
Kinetics
Isotherms
Mechanism

ABSTRACT

Excessive phosphate always leads to eutrophication in water bodies, which has been paid much attention in recent years. In this work, sewage sludge derived adsorbent was used to remove phosphate from aqueous solutions with the activation of $ZnSO_4 \cdot 7H_2O$ and H_2SO_4. The suitable dosages of the activation agents with the production of the carbonized sludge were determined. The adsorption performance was evaluated with different initial phosphate concentrations. The kinetics and isotherms were also discussed. Adsorbent characterization was studied by some analysis methods such as X-ray diffraction (XRD) and FTIR to understand the adsorption mechanism. In addition, the effects of pH, temperature, and adsorbent dosage on phosphate removal were investigated. The maximum phosphate adsorption capacity of the adsorbent was 123.46 mg g^{-1}. The sorption kinetic data had excellent fitness of the pseudo-second-order equation, indicating that the removal process belonged to chemisorption. Under the optimal condition (pH 5.0, temperature 35 °C, adsorbent 0.15 g), more than 99% of 65 mg l^{-1} initial phosphate were removed within 0.5 h. The results suggested that $ZnSO_4 \cdot 7H_2O$ and H_2SO_4 enhanced the phosphate adsorption and the adsorbent had the potential to remove phosphate effectively from aqueous solutions.

1. Introduction

Recent years have seen growing concerns in environmental pollution along with the rapid development of industry, which leads to the improvement of environmental technologies, materials as well as methods of waste disposal [1]. Wastewater treatment as one of the important aspects in environment protection can be carried out by physicochemical and biological methods [2]. There has been wide application of biological treatment processes which always have high removal efficiencies of pollutants as well as low cost [3]. However, a large amount of municipal sewage sludge is generated since they are inevitable by-products from numerous domestic and industrial wastewater treatment systems [4]. This kind of sludge always contains heavy metals, viruses as well as a variety of microorganisms [5]. Without appropriate disposal, surplus sludge will result in secondary pollution to soil, ground water and air [6]. Conventional treatments of sewage sludge are landfill, incineration, farmland utilization, etc. However, these options have severe drawbacks such as land occupation and pollutants emission, which prompt innovations for beneficial utilization of sewage sludge [7]. One approach to reuse the waste sludge is to convert sewage sludge into adsorbents. This method can not only reduce the amount of excess activated sludge but also achieve resource utilization, which meets both environmental and economic requirements and saves non-renewable natural resources [8].

The production of carbonized sludge has been popular in recent years. Sludge-derived char has been widely used as adsorbents for the treatment of effluents containing various pollutants such as dyes, heavy metals, and phenol [9–11]. Owing to its porous structure, this kind of material showed satisfactory adsorption performance. In order to improve adsorption capacity of sludge-based adsorbents, several methods have been used to change porosity as well as surface area or to modify surface groups [12]. Sludge-based adsorbents can be activated by physical or chemical processes. Physical activation is carried out by partial gasification agents such as steam, CO_2, and N_2, the products of which generally have relatively low surface areas [13,14]. For significantly improving adsorption capacity of the sludge-based materials, the carbonized sludge was prepared with several chemical agents. For instance, a kind of carbonized sludge supported by iron oxide was activated with $ZnCl_2$, which had high adsorption capacity and degradation efficiency when treating biologically pretreated coal gasification wastewater [15]. There were also several reports mentioning sludge carbonization activated with acid such as H_3PO_4 [16]. Although these chemical agents employed for sludge activation had different effects, the final products showed better removal ability of pollutants compared with physically activated adsorbents [17].

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http://dx.doi.org/10.1016/j.jtice.2017.04.002

Table 1
Chemical composition of dewatered sewage sludge.

Chemical composition	Concentration (wt%)	Chemical composition	Concentration (wt%)
C	39.05	SiO_2	35.9
O	29.24	Al_2O_3	11.9
N	0.77	Fe_2O_3	4.344
Al	6.297	P_2O_5	2.2
P	0.962	MgO	0.868
K	1.198	CaO	1.2
Fe	3.038		
Ca	0.86		

The discharge of effluents containing high concentrations of phosphorus has received considerable attention since excessive phosphorus causes eutrophication, which does great harm to ecosystems. In this work, a novel sludge-derived adsorbent was used with phosphate as adsorbates. The adsorption performance of the material was evaluated by the variation of several parameters such as initial phosphate concentration, pH value, temperature as well as dose of adsorbents. Moreover, the kinetics, isotherms and properties of adsorbents were investigated for a better understanding of the phosphate adsorption mechanism. The study was expected to provide a promising alternative in phosphate removal from aqueous solution.

2. Material and methods

2.1. Sludge and reagents

The activated sludge used in this study came from the Shahu municipal wastewater treatment plant in Wuhan, Hubei Province, China. The sewage sludge had been dewatered in the wastewater treatment plant before collection. The dewatered raw sludge was treated by using an oven at 105 °C for 48 h to make its weight constant. The principal chemical compositions of the dried sewage sludge are shown in Table 1. The chemical reagents used in this study such as $ZnSO_4{\cdot}7H_2O$, KH_2PO_4 were purchased from the National Medicines Corporation Ltd. of China.

2.2. Adsorbent preparation

After crushing of dried raw sludge, the carbonized sludge was prepared by using zinc sulfate. 10.0 g powder sludge was put into 75 ml deionized water supplied with $ZnSO_4{\cdot}7H_2O$ ranging from 0.05 to 0.25 mol at intervals of 0.05 mol. The mixture was stirred over 12 h to make it homogenous. Subsequently, the sol-gels were dried at 105 °C for several days until the weight was constant. The dried paste was then placed into a programmable electric furnace and heated for 2 h at 500 °C. The biomass was cooled to room temperature followed by ground to reach particle size less than 200 μm. In order to adjust the pH of the products to 7.0, the adsorbents were washed for several times with deionized water before dried at 105 °C. The final products activated by 0.05, 0.10, 0.15, 0.20, and 0.25 mol of $ZnSO_4{\cdot}7H_2O$ were labeled from CZ1 to CZ5 and tested as adsorbents to remove phosphate from aqueous solution aiming at obtaining the suitable dose of zinc sulfate.

For the sake of improving adsorption capacity of the sludge-based adsorbent, H_2SO_4 with the dose of 0.05, 0.10 and 0.15 mol was added in the sol-gels before mixed to homogenous paste by magnetic stirring apparatus. The other preparation procedures were the same as previous stage. After cooled, washed and dried, the three samples were used to adsorb phosphate and the optimal dose of H_2SO_4 was found. The final products were stored in an air-tight container for subsequent tests.

2.3. Adsorption tests

The phosphate adsorption tests were performed in 250 ml flasks. The removal performance of phosphate by carbonized sludge samples with different doses of the two chemicals during adsorbent preparation was evaluated using the following procedure: 0.1 g adsorbents were added in 100 ml aqueous solution with 50 mg l^{-1} of phosphate. The flasks were subsequently placed in a rotary shaker at 25 °C and 160 rpm for 24 h. The samples were collected during the adsorption process for measuring phosphate concentration. The amount of phosphate removed by the sludge-based char was calculated by

$$q = \frac{C_o - C_e}{m} \cdot V$$

where C_o is initial concentrations of adsorbate in contact with the adsorbent (mg l^{-1}), C_e is equilibrium concentrations of adsorbate in solution (mg l^{-1}), V is the volume of phosphate solution (L) and m is the mass of carbonized sludge.

In order to find the optimal parameters of the sludge-derived activated carbon, adsorption conditions were varied in batch tests. 0.1 g adsorbents were added in 100 ml aqueous solution with different phosphate concentration ranging from 50 mg l^{-1} to 330 mg l^{-1}. The flasks were shaken at 25 °C until the phosphate concentration reached equilibrium. The results were used for fitting isotherms as well as kinetic models. Batch experiments at different pH values were carried out by adjusting the initial pH of the solution in five flasks to 3.0, 5.0, 7.0, 9.0, 11.0, respectively, with supplement of 1 M NaOH and 1 M HCl solution. The effects of temperature and adsorbent dose were investigated by the variation of temperature as well as mass of carbonized sludge ranging from 15 to 45 °C and 0.06 to 0.15 g, respectively. The mixtures were shaken at the speed of 160 rpm during the tests and samples were collected periodically.

2.4. Analytical methods

During the whole experiment, samples were obtained at intervals of 0.5 h. The mixtures were taken from the flasks and filtered immediately by using a 0.45 μm filter. The concentration of phosphate was measured according to the standard methods [18]. The pH values of wastewater in the system were monitored by a pH meter (PHS-3C). All samples were analyzed in triplicate. The analysis of chemical compositions of dried sewage sludge was performed by using X-ray fluorescence spectrometer (XRF, S4 Pioneer, Bruker AXS, Germany). The morphologies of the sludge char in this study were observed by using a scanning electron microscope (SEM, VEGA 3 LMU, TESCAN, Czech Republic). The surface functional groups of the carbonized sludge were determined by a Nicolet 5700 FTIR spectrometer in the range of 4000–400 cm^{-1}. An X-ray diffractometer (X' Pert Pro, PANalytical, Netherlands) was used to record the XRD patterns of the adsorbent samples in the 2θ range of 10–80°.

3. Results and discussion

3.1. Removal performance of phosphate during adsorbent preparation

The adsorption performance of phosphate by different adsorbent samples was investigated and the time courses were shown in Fig. 1. It can be seen from Fig. 1(a) that the phosphate removal rate increased with higher concentration of zinc sulfate. Within the first hour, the concentration of phosphate decreased from 50.19 to 45.24 mg l^{-1} by using CZ1 while the phosphate concentration reduced to 35.59 mg l^{-1} when adding CZ2 to the solution. The removal efficiencies of phosphate were 33.90%, 65.55%, and 80.39%,

Please cite this article as: Y. Jiang et al., Removal performance of phosphate from aqueous solution using a high-capacity sewage sludge-based adsorbent, Journal of the Taiwan Institute of Chemical Engineers (2017), http://dx.doi.org/10.1016/j.jtice.2017.04.002