Journal of Cleaner Production 175 (2018) 572–581

Contents lists available at ScienceDirect

Journal of Cleaner Production

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

Preparation, optimization, and application of sustainable ceramsite substrate from coal fly ash/waterworks sludge/oyster shell for phosphorus immobilization in constructed wetlands

Gong Cheng [a], Qionghui Li [b], Zhan Su [c], Sheng Sheng [c], Jie Fu [d], *

[a] *Shenzhen Academy of Environmental Sciences, Shenzhen 518001, China*
[b] *School of Civil and Environmental Engineering, Shenzhen Graduate School, Harbin Institute of Technology, Shenzhen 518000, China*
[c] *Huadong Engineering Corporation Limited, Hangzhou 311122, China*
[d] *Department of Environmental Science & Engineering, Fudan University, Shanghai 200433, China*

ARTICLE INFO

Article history:
Received 2 September 2017
Received in revised form
10 December 2017
Accepted 11 December 2017
Available online 14 December 2017

Keywords:
Constructed wetlands
Phosphorus adsorption
Ceramsite
Coal fly ash
Waterworks sludge

ABSTRACT

This study has prepared a sustainable and efficient ceramsite substrate for phosphorus immobilization in constructed wetlands by adopting coal fly ash (CFA) and waterworks sludge (WS) as the main materials, and oyster shell (OS) as the additive. The optimal parameters for CFA/WS/OS ceramsite preparation were determined as follows: mass ratio of CFA:WS:OS = 6:4:0.8, preheating temperature = 600 °C, preheating time = 30 min, calcining temperature = 1050 °C, and calcining time = 8 min. The CFA/WS/OS-op ceramsite, obtained under the optimal condition, had a microporous structure with crystal strength components and active calcium components. Static adsorption experiments indicated neutral-alkaline conditions and/or higher temperature were conducive to improve the phosphorus removal by CFA/WS/OS-op; the adsorption kinetics and isotherm could be well described by the second-order model and Langmuir model, respectively. The theoretical maximum phosphorus adsorption capacity of CFA/WS/OS-op at 35 °C reached up to 4.51 mg/g. Speciation analysis on the adsorbed phosphorus revealed chemisorption by calcium in CFA/WS/OS-op was the dominant adsorption mechanism. In dynamic column experiments, as much as 90% of the phosphorus in raw water collected from the secondary effluent of a sewage treatment plant was removed by CFA/WS/OS-op ceramsite with a hydraulic retention time of 12 h.

1. Introduction

Eutrophication problems, caused by the massive use of fertilizers and the increased production of wastewater, are besetting territories with intensive agricultural, urban, and/or industrial activities (Tercero et al., 2017; Wang et al., 2015). Although phosphorus (P) is an indispensable macronutrient in aquatic environments and highly required for the growth of living organisms and the normal functioning of ecosystems (Xiong et al., 2017), excessive phosphorus has been recognized as the major factor to trigger the eutrophication effect, water quality deterioration, and ecosystem degradation (Ngatia et al., 2017; Peng et al., 2012). It is estimated that about 1.3 million tons of phosphorus is discharged into aquatic systems every year from all over the world (Xiong et al., 2017). Therefore, the control of phosphorus loads to waterbodies has become an important aspect of water environment governance.

Recently, constructed wetlands (CWs) are increasingly being recognized as a relatively low-cost, energy-efficient, natural, and ecological technology for treating municipal and/or domestic wastewaters (Luo et al., 2017a), which have been designed and constructed to utilize the natural purification processes involving wetland plants, substrates, and the associated microbial assemblages (Gao et al., 2017). CWs are broadly approved for the ability to remove phosphorus through interactions among substrates, plants, and microorganisms (Gao et al., 2017; Luo et al., 2017a; Yin et al., 2017). Adsorption and precipitation by the substrate in CWs are the dominant mechanisms for phosphorus removal, which are especially evident in saturated subsurface flow CWs (Bowden et al., 2009; Kaasik et al., 2008). Since the substrate largely determines the phosphorus removal efficiency of CWs (Hua et al., 2016), selection of the proper substrate is very important in the construction of CWs.

---

* Corresponding author.
E-mail address: jiefu@fudan.edu.cn (J. Fu).

https://doi.org/10.1016/j.jclepro.2017.12.102
0959-6526/© 2017 Elsevier Ltd. All rights reserved.

Traditionally, some inert materials such as sand, unpolluted soil, and gravel are used as the substrates in CWs (Brix et al., 2001), however, the low adsorption capacity of these materials always affects the nutrients removal efficiency (GarcÍA et al., 2010). Recently, many active materials containing abundant Al, Ca, Fe, or Mg, which have a high affinity for phosphorus and low economic costs, have been selected as the substrates in CWs, and this practice has gained lots of attentions (Claveau-Mallet et al., 2012; Yin et al., 2017). To date, a large number of active materials have been used and/or proposed as substrates in CWs, including natural materials (mineral, rock, and oyster shell), industrial byproducts (slag, coal fly ash, and waterworks sludge), and man-made products (lightweight aggregate, polonite, and ceramsite) (Huang et al., 2017; Mann, 1997; Vohla et al., 2011; Wu et al., 2016). However, the direct use of raw materials will generate some problems, such as relatively low adsorption capacity, secondary pollution, low stability and mechanical strength (Tang et al., 2014; Yu et al., 2010). Ceramic production of raw materials by high temperature calcination is a promising method to improve the material properties and safety, and extend their applications.

Ceramic production can provide many benefits to the products such as improving mechanical strength by solidification/stabilization, removing the organic pollutants/pathogenic agents by hyperthermal decomposition and gasification, stabilizing the harmful metals by incorporating into crystal phases, and enhancing the adsorption capacity by generation of microporous structures (Tang et al., 2014; Xu et al., 2008; Yu et al., 2010). Coal fly ash (CFA) and waterworks sludge (WS) are two industrial byproducts with a high adsorption capacity. The large contents of SiO_2 and Al_2O_3 in CFA and WS make them fit as ceramic skeleton components (Lu et al., 2013; Luo et al., 2017b). Previous studies have mainly focused on the ceramic production using unitary fly ash or sludge as the main raw material. Only two studies reported preparing the ceramiste by using binary main raw materials with fly ash and sludge. However, one study (Yang et al., 2013) used the river silt, and the other (Shao et al., 2012) employed the sewage sludge, which has a high content of volatiles relative to WS and cannot provide high strength ceramic products (Xu et al., 2008). Moreover, the aforementioned two studies mainly focused on the preparation of fly ash/sludge ceramiste and the ceramic applications like adsorbing phosphorus have not been addressed. To date the preparation and application of CFA/WS ceramsite as the substrate in CWs have not been reported yet. The composition difference between CFA and WS may have a complementary effect during the sintering process. For example, the high content of Al_2O_3 in WS (Luo et al., 2008) may promote the formation of high strength mullite, while the relatively high content of fluxing components (e.g., CaO, Fe_2O_3, MgO, and SO_3) in CFA (Zhao et al., 1997) may reduce the sintering temperature and improve the sintering performance. Yet, these hypothetic effects need the experimental proof.

In addition, in order to improve the expansion and porosity of ceramsite, some components of producing gas at the high temperature are needed during the sintering process, such as iron oxides, carbons, and carbonates (Reed and JamesStalford, 1988). Oyster shell (OS) is a natural waste and exhibits a good adsorption capacity for phosphorus (Park and Polprasert, 2008). The main component of OS is $CaCO_3$ (>95% by weight) (Lee et al., 2008), which can be decomposed to CaO and CO_2 at a high calcining temperature. The generated CaO can work as the fluxing agent to reduce the activation energy of ceramic melt, and CO_2 gas can perform as the bulking agent to increase the porosity of ceramsite. Therefore, OS may be used as a functional additive to improve the sintering process and properties of CFA/WS ceramsite. However, employing OS into the raw materials for manufacture of ceramsite has not been reported yet, and the feasibility and process for the production of ceramsite by adopting OS additive have been kept unexplored.

This study first synthesized CFA/WS ceramsite and investigated the preparation parameters via an orthogonal experimental design. Oyster shell was then added to the formula as a functional additive and the optimal parameters were determined by a one-factor experimental design. The phosphorus adsorption capacity and mechanism of CFA/WS/OS ceramsite were explored successively by static adsorption experiments and dynamic column experiments (Scheme 1). The purpose of this study is to develop a stable ceramsite substrate for CWs with a good phosphorus adsorption capacity, which can improve the phosphorus immobilization in CWs and extend the substrate lifetime.

2. Materials and methods

2.1. Materials

All chemicals used in this study were of analytical grade or better. Coal fly ash was collected from Pinghai Coal Power Plant (Huizhou, Guangdong, China). Waterworks sludge (moisture content = 80%) was obtained from Buji Water Plant (Shenzhen, Guangdong, China). Oyster shell was purchased from Tiechong Oyster Farm (Huizhou, Guangdong, China).

2.2. Preparation of ceramsites

Raw materials (i.e., CFA, WS, and OS) were dried at 105 °C for 4 h and passed through a 120-mesh sieve. A mixture of raw materials with different mass ratios was mixed evenly with water (30–40 wt %), rubbed into pellets (6–8 mm), and dried at 105 °C for 2 h. The ceramic blank pellets were then subjected to preheating at 300–600 °C for 15–30 min and calcining at 1000–1150 °C for 5–30 min successively with an SX2-5-12 box-type resistance furnace (Yiheng Scientific Instrument Co., Ltd., Shanghai, China). After cooling, the finished ceramsite was gained.

2.3. Material characterization

Elemental analysis was determined by X-ray fluorescence spectrometry (XRF) using an XRF-1800 system (Shimadzu, Tokyo, Japan). The X-ray diffraction (XRD) measurements were performed on an X'Pert PRO MRD/XL system (Panalytical, Almelo, the Netherlands). Scanning electron microscope (SEM) images were obtained with a VP-FE-SEM system (Carl Zeiss, Oberkochen, Germany). Thermogravimetry-differential scanning calorimetry (TG-DSC) was conducted on an STA449F3 instrument (Netzsch, Selb, Germany). The bulk density and apparent density of ceramsite were measured according to GB/T 17431.1–2010 standard method (2010). The compressive strength of ceramsite was determined by reported method (Li et al., 2000). The heavy metal leaching toxicity analysis was conducted by HJ/T 299–2007 method (2007b), and GB 5085.3–2007 standard (2007a) and GB 3838-2002 standard (2002b) were employed as the evaluation criteria.

2.4. Static adsorption experiments

All the static adsorption experiments were carried out in glass Erlenmeyer flasks (total volume = 100 mL) with 50 mL of KH_2PO_4 solution and the ceramsite on an SHA-C thermostatic oscillator (Jintan Medical Instrument, Jintan, Jiangsu, China) with a speed of 100 rpm. For kinetic studies, 1 g of ceramsite was added into KH_2PO_4 solution (total phosphorus (TP) = 50 mg/L). After the pH value was adjusted to 7 using dilute HCl and NaOH aqueous solution (aq.), the mixture was shaken at 30 °C for different times