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Immobilization of heavy metals in ceramsite produced from sewage sludge biochar

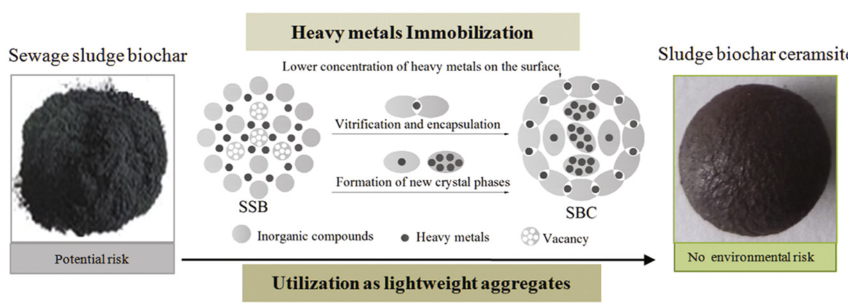
Jie Li [a,b], Guangwei Yu [a,*,1], Shengyu Xie [a,b], Lanjia Pan [a,b], Chunxing Li [a], Futian You [a], Yin Wang [a,*]

[a] Key Laboratory of Urban Pollutant Conversion, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China

HIGHLIGHTS

- The SBC was successfully produced using SSB directly.
- The HMs were well immobilized in SBC without environmental risk.
- HMs immobilization was mainly related to formation of new crystal phases.

GRAPHICAL ABSTRACT



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ABSTRACT

Ceramsite was prepared from sewage sludge biochar (SSB). The migration, speciation evolution, leaching toxicity, and potential environmental risk of heavy metals (HMs) in sludge biochar ceramsite (SBC) were investigated. The characteristics of the SBC met the requirements for Chinese lightweight aggregate standards (GB/T 1743.1-2010 and JT/T 770-2009) and the heavy metals (HMs: Cu, Zn, Cr, Pb, and Cd) were well immobilized in the SBC. The leaching percentages of the HMs in SBC were remarkably reduced, in particular after preheating at 400 °C and sintering at 1100 °C. The leaching percentages of Cu, Zn, Cr, Cd, and Pb decreased from (19.099, 18.009, 0.010, 3.952, and 0.379) % to (2.122, 4.102, 0.002, 1.738, and 0.323) %, respectively. The RAC values of the HMs in SBC were all lower than 1%, and the risk index (*RI*) suggested that the SBC had no HMs contamination and very low potential ecological risk when used in the environment. Furthermore, the HM-immobilization mechanisms were mainly related to the formation of new crystal phases (silicate and phosphate minerals) by incorporation of HMs, and to vitrification and encapsulation with low concentration of HMs on the surface. This work provides a useful method for large-scale reuse of SSB with very low leaching toxicity and low potential ecological risk of HMs.

1. Introduction

With the rapid development of urbanization, large amounts of sewage sludge (SS) are generated from wastewater treatment plants every year. In China, >60 Mt of municipal sewage sludge (80% water content) have been produced since 2015 (Jin et al., 2016). In addition, public consciousness has motivated the development of new environment-friendly technologies to minimize the environmental impact of SS. Sludge pyrolysis/carbonization, a thermal treatment process in an inert gas, has proven to be a promising technology for the treatment of SS. It can eliminate almost all the pathogens and organic pollutants in SS, and the volume of the SS is significantly reduced after pyrolysis,

* Corresponding authors.
E-mail addresses: gwyu@iue.ac.cn (G. Yu), yinwang@iue.ac.cn (Y. Wang).
[1] Co-first author.

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with biochar remaining (Dou et al., 2017). An SS treatment technology combining hydrothermal treatment and pyrolysis has been developed, and a pilot-scale plant for pyrolysis of hydrothermally treated SS (capacity 30 t-SS per day) was established in Xiamen by the Institute of Urban Environment of the Chinese Academy of Sciences (J. Li et al., 2017; C.X. Li et al., 2017). For the application of sewage sludge biochar (SSB), the prevalent approach to disposal of SSB is to use it as fertilizer in soil, and the effect of SSB on the migration of heavy metals (HMs) in soil was investigated (Khan et al., 2013). However, our previous study showed that the HMs in SS are enriched during the process of hydrothermal treatment and pyrolysis (Wang et al., 2016), and that the large-scale utilization of SSB is limited by the high concentration of HMs in SSB (Lucchini et al., 2014; Van Wesenbeeck et al., 2014). The HMs in SSB can migrate through the action of plants and microbes, which could cause potential soil and water contamination. Therefore, there is an urgent need to find other feasible methods for the large-scale utilization of SSB.

Ceramics is considered to be a promising and widely applied approach for treating a variety of hazardous wastes including sludge, slurries, contaminated soils, dust, and other particulate matter. It has been reported that SS can be used to produce ceramics (Kizinievič et al., 2016; Xu et al., 2008b, 2010). However, producing ceramsite using SS directly has some shortcomings: (1) the composition of sludge is complex, and includes pathogens and organic pollutants that can be transformed into gaseous pollutants (dioxin and polycyclic aromatic hydrocarbon) during the sintering process. (2) The cost is high and the transportation challenging for such enormous volumes of SS with high water content.

The SSB obtained from pyrolysis of SS, possesses less volume, water, and organic pollutants. More importantly, the components of SSB are mainly SiO_2, Al_2O_3, Fe_2O_3, CaO + MgO, and K_2O + Na_2O, which play significant roles in the production of ceramsite materials (J. Li et al., 2017a). Thus, the specific properties of SSB make it easy to utilize as feedstock for production of ceramsite. Furthermore, the high temperature used to produce ceramsite can trap the HMs in the durable metallic mineral phase (Xiao et al., 2015; Xu et al., 2010). In addition, the ceramsite materials usually exhibit high permeability, resistance to change by heat, porosity, specific surface area, and chemical resistance. These characteristics have resulted in ceramsite being widely used as filter material for environmental protection or as construction materials such as lightweight aggregates (Han et al., 2011; Qiu et al., 2010; Xu et al., 2008a).

To utilize SSB on a large scale, a process for preparation of ceramsite using SSB was developed and the feasibility of ceramsite as a lightweight aggregate was evaluated according to the Chinese standards for lightweight aggregates (GB/T 1743.1-2010 and JT/T 770-2009). Furthermore, the migration, speciation evolution, leaching toxicity, and potential environmental risks of HMs were investigated. The immobilization mechanism of HMs in SBC was explored by scanning electron microscopy (SEM), energy dispersive spectrum (EDS), and X-ray diffraction analysis (XRD).

2. Materials and methods

2.1. Materials

The SSB used in this study was obtained from the pilot-scale plant established by the Institute of Urban Environment in a wastewater treatment plant at Xiamen, Fujian Province, China. The SSB was pyrolyzed at 600 °C for 30 min in a rotary furnace from dewatered sludge that was obtained after previous hydrothermal treatment (180 °C for 30 min). The ultimate and proximate analyses and main chemical components of SSB are shown in Table 1.

Table 1
Ultimate and proximate analyses and main chemical components of SSB.

Items		Values	Items		Values
Proximate analysis (wt%)[a]	Ash	82.65	Main chemical components of ash (wt%)	SiO_2	23.57
	VM	5.62		Al_2O_3	15.94
	FC	11.73		Fe_2O_3	19.86
Ultimate analysis (wt%)[a]	C	8.21		MgO	8.55
	H	0.21		CaO	6.87
	N	0.30		Na_2O	1.59
	O	8.22		K_2O	1.05
	S	0.41		Other	5.22

[a] On dry weight basis; O = 100 − (C + H + N + S + Ash).

2.2. Methods

2.2.1. Preparation process of SBC

The SSB was ground and passed through a 100-mesh sieve before being mixed with 35 wt% water. The mixture was stirred and poured into a pelletizer to make raw pellets with diameter of 10 mm. The raw pellets were preheated and sintered after being dried at 105 °C in an air oven for 3 h (Fig. 1). To determine the effects of preheating and sintering temperatures on the immobilization of HMs, the raw pellets were sintered at three targeted temperatures (1000, 1050, and 1100 °C) after being preheated at 400 °C (designated as SBC400–1000, SBC400–1050, and SBC400–1100), and the raw pellets were preheated at three preheating temperatures (300, 400, and 500 °C) before being sintered at 1050 °C (designated as SBC300–1050, SBC400–1050, and SBC500–1050). The dwelling time of preheating and sintering were both 30 min with a ramp rate of 10 $°C\cdot min^{-1}$ during the whole calcination process. Overall, five kinds of SBC samples were obtained after being cooled to 30 °C in the furnace.

2.2.2. Speciation and residual analysis of HMs

The speciation of HMs (Cu, Zn, Cr, Pb, and Cd) in the samples was determined through the three-step sequential extraction procedure proposed by the European Community Bureau of Reference (BCR) (Baig et al., 2009). The acid soluble/exchangeable fraction (F1), reducible fraction (F2), and oxidizable fraction (F3) were extracted according to the literature (Baig et al., 2009). The residual fraction (F4) and the total concentration of HMs were detected after digestion *via* microwave in an acid mixture (HNO_3:$HClO_4$:HF = 5:5:2, v/v/v) (Wang et al., 2016).

All the extracted samples were passed through 0.45 μm membrane filters before being diluted to a constant volume with 2% HNO_3 and stored in a refrigerator at 4 °C until analysis. The concentrations of Cu, Zn, Cr, Pb, and Cd in the samples were measured by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500cx, Agilent, USA). Quality assurance and quality control (QA/QC) for the metals analyses were performed using duplicates, method blanks, standard reference materials, and GSS-1 soils, obtained from the Center of National Standard Reference Material of China. All of the measurements were repeated three times, and their average values were used in the data analyses.

The residual rate (R, %), the percentage of each target heavy metal remaining in the SBC, was determined as follows (Chen and Yan, 2012):

$$R = x_2 \cdot m_2 / (x_1 \cdot m_1) \cdot 100 \tag{1}$$

where x_1 is the concentration of the target heavy metal in SSB ($mg\cdot kg^{-1}$), x_2 is the concentration of the target heavy metal in SBC ($mg\cdot kg^{-1}$), and m_1 and m_2 are the masses of the SSB and SBC, respectively (kg).

2.2.3. Leaching toxicity analysis of HMs

The leaching toxicity characteristics of HMs were determined by the toxicity characteristic leaching procedure (TCLP). This is a standard method updated by the USEPA for determining leaching toxicity (Xu et al., 2010). This method has been widely applied for assessing the