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Improved utilization of phosphorous from sewage sludge (as Fertilizer) after treatment by Low-Temperature combustion

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ABSTRACT

The utilization of phosphorus from sewage sludge is an important method used to solve the shortage of phosphorus resources in the world. However, high levels of toxic compounds and low phosphorus bioavailability in sewage sludge are the main factors limiting its direct agricultural use. This paper proposes a low-temperature combustion method that can enrich the phosphorus in sludge ash. Low temperature-treated sewage sludge ash (LTSA) at different oxygen concentrations (20%, 60%, 100%) were obtained through a specific experimental device. Then, the species and leaching characteristics of phosphorus in LTSAs were analyzed and compared with pyrolysis sewage sludge char (PSSC) and incinerated sewage sludge ash (ISSA). Results show that low-temperature combustion of sludge increased the total phosphorus content in the bottom ash by 45.6%, and the bioavailable phosphorus content increased 2.9 times. Further, by increasing the concentration of oxygen while carrying out low-temperature combustion of sludge, part of the non-apatite inorganic P was converted to apatite P (AP), resulting in a 46.3% increase in AP in the sludge. Low-temperature combustion can also convert heavy metals (Cd, Cr, Cu, Pb, and Zn) in the sludge from an easily leachable form (acid extractable fraction and reducible fraction) to a stable form (reducible fraction) and decrease the leaching of heavy metals. Leaching of Cr and Cu decreased by 97.56% and 98.52%, respectively.

1. Introduction

Phosphorus (P) is an essential element in all organic life and it is fundamental in fertilizers used for food production. According to the International Water Management Institute, by the year 2050 the demand for phosphorus will grow by 70% overall and even by up to 100% in developing countries (Guedes et al., 2014). However, P is a non-renewable resource and the main source of phosphate is phosphate rocks. Many scientists claim that the phosphate rock deposits will be exhausted as soon as 100–130 years from now (Li et al., 2014). For these reasons, the usage of secondary raw materials has received increased to attention. Phosphorus is recovered from phosphorus-rich residues such as manure, meat, bone meal, and agricultural waste, even as sewage sludge can be part of the solution. (Azuara et al., 2013; BiplobKumarBiswas et al., 2009; Coutand et al., 2008; De Vries et al., 2012; Jin et al., 2009; Kaikake et al., 2009; Sano et al., 2012; Tan and Lagerkvist, 2011; Wzorek et al., 2006; Zhang et al., 2001).

After the process of urban sewage treatment, a large amount of phosphorus is concentrated in the residual sludge. The phosphorus content of residual sewage sludge used in the biological phosphorus removal process in modern wastewater treatment systems is about 4–9% (BiplobKumarBiswas et al., 2009). Thus, residual sewage sludge can be considered a promising source of phosphorus (Havukainen et al., 2016). However, sewage sludge contains toxic organic compounds such as polyaromatic hydrocarbon, chlorobenzene, and plasticizers, as well as pathogens, and heavy metals. (Carbonell et al., 2009; Grøn, 2007; Harrison et al., 2006). As a result, sewage sludge cannot be applied directly to the agricultural land as phosphate fertilizer.

Thermal treatment of sewage sludge can reduce waste volume, decompose organic pollutants, reuse the energy contained in the sludge, and generate valuable byproducts. It is a sustainable strategy for the disposal of sewage sludge (Hartman et al., 2005; Hossain et al., 2011). During the thermal treatment of sewage sludge, P was enriched in ash, which has several consequences (Moench, 2000; Hossain et al., 2011; Wang et al., 2012). Presently, the thermal treatment of sewage sludge mainly includes incinera-

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tion, pyrolysis, gasification, and wet oxidation. Thermal processing modifies the chemical and physical structure of the feedstocks, which affect P speciation, resulting in P-bioavailability varying between thermally processed materials (Cabeza et al., 2011; Hogan et al., 2001; Schiemenz and Eichler-Löbermann, 2010).

According to the result of Moench, 2000, P is a typical lithophilic element with approximately 80–90% of P remaining in the bottom ash after incineration. The rest is transferred into fly ash, mainly from mechanical carryover. However, during the process of sewage sludge incineration, a large amount of fly ash is produced (Lin et al., 2010), which reduces the amount of phosphorus in the bottom ash. Fly ash is especially problematic because of the high content of heavy metals that are easily released (Li et al., 2017), which makes it difficult to use the phosphorus in fly ash directly (Donatello and Cheeseman, 2013; Petzet et al., 2012). Therefore, to improve the utilization of phosphorus in the bottom ash, it is necessary to reduce the amount of fly ash produced during combustion and the phosphorus concentration in the fly ash. Fixed bed heat treatment technology is widely used in sludge pyrolysis and gasification technology (Wang et al., 2016; Werle, 2014; Werle and Wilk, 2012). One of its characteristics is that the amount of fly ash produced during the reaction is extremely small. The self-sustained combustion of sludge in a fixed bed under low temperature conditions can reduce the production of fly ash while utilizing the heating value of the sludge itself, and avoiding the risk of phosphorus volatilization caused by high temperature during incineration. This achieves the purpose of enriching phosphorus in sludge ash.

There has been much interest in the recovery of P from incinerated sewage sludge ash either as phosphoric acid or fertilizer (Adam et al., 2009; Atienzamartínez et al., 2014; Azuara et al., 2013; Cohen, 2009; Donatello et al., 2010; Franz, 2008; Ottosen et al., 2013). However, more research is needed. In particular, there are not any studies on the migration and transformation of P and heavy metals in sewage sludge during low-temperature combustion. The present article focusses on P-bioavailability and the migration and transformation of P in low-temperature combustion sewage sludge ash (LTSA). The chemical properties of P contained in LTSA, pyrolysis sewage sludge char (PSSC), and incinerated sewage sludge ash (ISSA) are compared and discussed in this article.

2. Materials and methods

2.1. Sewage sludge

Dewatered sewage sludge (moisture 76.5%) was sourced from a power plant in Zhejiang, China. More than 10.0 kg of sludge was dried at 102–105 °C in a constant-temperature oven overnight. Then, dried sewage sludge (DSS) was pulverized and passed through a 60-mesh (250 μm) sieve, sealed in a plastic zip-lock bag, then stored in a freezer until analysis. The moisture, ash, volatiles and fixed carbon were measured by the ASTM D5142 standard. Ultimate analysis was performed with an elemental analyzer 5E-CHN2000. The higher heating value was measured by the ASTM D3286-96 standard. The elements (except C, N, O, H) of the sewage sludge were analyzed by X-ray fluorescence (XRF).

2.2. Thermal treatments of sewage sludge

The schematic of the experimental apparatus setup for the low-temperature combustion of sewage sludge is shown in Fig. 1. The thermocouple group in Fig. 1 is numbered 1–5 from bottom to top. The interval between each thermocouple is 40 mm. After the device is filled with DSS, the heating plate is turned on, and the initial ignition position is adjusted by controlling the heating time.

Oxygen and nitrogen are introduced before the heating plate heats up, and the oxygen flow rate is controlled between 36 and 72 $L{\cdot}h^{-1}$. The proportion of oxidant gas is controlled by adjusting the flow rate of nitrogen and oxygen to provide the required amount of oxygen for low-temperature combustion. PSSC samples were produced by pyrolysis in an N_2 atmosphere (gas flow rate of 15 $L{\cdot}h^{-1}$) in a fixed bed horizontal tube furnace. Twenty grams of DSS were fed into the reactor each time and pyrolyzed at temperatures of 400 °C, 600 °C, and 800 °C. The three series of ISSA were obtained by combustion of the sludge in air in a self-made small fluidized bed reactor at different temperatures (400 °C, 600 °C, and 800 °C). The residue was collected and stored in zip-lock bags.

2.3. The bioavailability of phosphorus

The P-bioavailability of the products was analyzed by using neutral ammonia citrate (NAC), 2% citric acid (CA) (Steckenmesser et al., 2017; Wang et al., 2012). These extraction tests were carried out according to the EU regulation No. 2003/2003 (EU. 2003). The P-concentration of the solutions was determined afterwards with the molybdenum blue method.

2.4. The speciation of phosphorus

The Standards, Measurements, and Testing (SMT) harmonized procedure was adopted for the determination of the P speciation. The SMT protocol is a modified version to accommodate all five types of P speciation: non-apatite inorganic P (NAIP; the forms associated with oxides and hydroxides of Al, Fe, and Mn), apatite P (AP; the forms associated with Ca), inorganic P (IP), organic P (OP), and total P (TP) (Pardo et al., 2003; Ruban et al., 2001). The outline of the SMT protocol and the details of the analytical steps are presented in Ruban et al., 2001 and Medeiros et al., 2005. The validity of the SMT protocol and the accuracy with which it has been carried out may be tested by comparing the sum of the single speciation with the TP measured separately. That is TP = IP + OP and IP = NAIP + AP. The P-concentration in the solutions was determined afterwards with the molybdenum blue method and a spectrometer.

2.5. Leaching of heavy metals

The toxicity characteristic leaching procedure (TCLP) is a standard extraction test designed by the US Environmental Protection Agency (EPA) to evaluate the leaching potential of heavy metals in solid waste. The sample was preliminarily evaluated to determine which extraction fluid (solution #1 pH 4.9 ± 0.05, solution #2 pH 2.88 ± 0.05) was more appropriate for the products. The liquid to solid ratio was 20 mL/1 g and the mixture was agitated for 18 h with a rotary tumbler at 30 ± 2 rpm. The leachate was separated from the solid phases by filtering through the glass fiber filter paper and then was acidified using 2 ml nitric acid per 100 ml leachate before being analyzed by ICP-AES (thermo, iCAP 6300).

Sequential extraction of heavy metals: Heavy metals fractions in the sewage sludge were determined by the European Community Bureau of Reference (BCR) method. Sequential extraction was performed using a three-stage modified procedure recommended by BCR and analyzing the various solutions and the solid residue. (Table 1).

Acid extractable fraction: 1.0 g LTSA sample was extracted with 40 mL of 0.11 $mol{\cdot}L^{-1}$ acetic acid solution by shaking in a mechanical, end-over-end shaker at 30 ± 10 rpm at 22 ± 5 °C for 16 h. The extract was separated by centrifugation at 3000 rpm for 20 min, collected in polyethylene bottles and stored at 4 °C until analysis. The residue was washed by shaking for 15 min with 20 mL of dou-