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Comparative study on the mobility and speciation of heavy metals in ashes from co-combustion of sewage sludge/dredged sludge and rice husk

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HIGHLIGHTS

- Co-combustion of rice husk and dredged/sewage sludge.
- Basic performance and environment property of by-product from co-combustion.
- Multi-step leaching was designed to test the mobility of heavy metals (Cr, Cu, Fe, Mn, Ba and Zn).
- Combining leachability and speciation to assess the environmental risk of the ash.

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ABSTRACT

The co-combustion of sludge (sewage and dredged sludge) with rice husk is expected to become a trend because of its economic and environmental benefits. However, the massive residues from the co-combustion process and the mobility of heavy metals (HMs) warrant special attention. The basic performance and environmental properties of the trace elements (Cr, Cu, Fe, Mn, Ba and Zn) from the co-combustion ashes were studied to promote the further utilization of these materials. These ashes have a shell particle shape, high specific area, high amorphous content and low crystalline phase content. The investigation mainly focused on the environmental properties of these ashes to evaluate the risk of these by-products to the environment. Results show Cu, Mn, and Zn have cumulative leaching concentrations of 1.033, 23.32, and 3.363 mg/L for W, by contrast, Cr, Cu, Fe, Mn, Ba, and Zn have cumulative leaching concentrations of 0.488, 0.296, 8.069, 10.44, 2.568, and 2.691 mg/L for H, which are much greater than the Chinese ground water standard (GB/T14848-93). Meanwhile Mn, Zn, Ba, Cr, and Fe all pose a very high risk for H, while Cu only poses a medium risk, and all HMs in W exhibit much lower contamination levels than those in H by the method of risk assessment code (RAC). It indicates that these ashes have undesirably high levels of HMs that demonstrate high mobility and pose environmental risks according to their leachability and chemical speciation. And the HMs in W show lower mobility and environmental hazards than those in H.

1. Introduction

The high demand for excellent water quality and the strict environmental laws and regulations have sharply increased the amount of sewage sludge produced from wastewater disposal (Werle and Wilk, 2010). Sewage sludge is a complex heterogeneous mixture of organic compounds and inorganic matter (Chen et al., 2008; Pettersson et al., 2008a,b) that can be used as a source of renewable energy. Contamination by heavy metals (HMs) is among the most serious drawbacks in the use of sewage sludge (Cai et al., 2007). However, HMs cannot be degraded or physically neutralized.

Sediments have become a sink and source of toxic components because of the gathering and resuspension of various pollutants. Therefore, sediments pose serious threats to water quality (Bastami

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et al., 2012; Knox et al., 2016). Plenty of sediments are being dredged every year to relieve their accumulation (Ho et al., 2012). However, the presence of HMs in dredged sludge restricts the reuse of this material (Perelo, 2010).

Given these facts, the appropriate treatment and disposal of sludge (sewage and dredged sludge) have recently gained increasing attention. Co-combustion is currently being seen as an economic solution to sludge treatment (Otero et al., 2008; Rui et al., 2009; Calvo et al., 2013; Magdziarz and Wilk, 2013; Agbor et al., 2014). However the co-combustion of sludge and fossil fuels may emit large amounts of carbon dioxide and other pollutants. Replacing fossil fuels with another green energy source presents a viable solution to this problem. Among the available green energy sources, rice husk offers an ideal option because of its high calorific value and abundant supply. Therefore, to achieve sustainable electricity production and reduce pollution, blending the fuels of sludge and rice husk under well-controlled conditions is expected to become a trend in the future.

Although co-combustion can reduce toxic organics pollutants, the HM pollution resulting from this process remains unresolved (Lopes et al., 2005; Manskinen et al., 2011; Freire et al., 2015). Therefore, evaluating such pollution is essential in the development of an efficient co-combustion technology. Many studies (Gulyurtlu et al., 2006; Van Loo and Koppejan, 2008; Rui et al., 2009) show that HMs mostly accumulate in ashes during the co-combustion process. Moreover, these ashes have unstable chemical characteristics depending on the type of fuel (Hupa, 2005; Kalembkiewicz and Chmielarz, 2012). Therefore, the ashes produced during the co-combustion of sludge and rice husk must be thoroughly examined.

The transition from the stabilized ash to the liquid medium represents the main release mechanism of HMs to the environment. Therefore, assessing the environmental performance of a by-product based on its total HMs content may yield to unreliable results and overestimate the related environmental risks, thereby limiting the application of co-combustion ashes. Accordingly, the characterization and mobility of HMs has received considerable research attention. The leaching behavior of co-combustion ashes (forest biomass and sewage sludge) has been extensively characterized in previous research (Pettersson et al., 2008a,b; Calvo et al., 2013). However, the leaching properties of co-combustion ashes may significantly differ from the co-firing of sludge and rice husk. Many leaching tests are conducted with different reagents, temperatures, agitation methods, liquid–solid ratios and contact times for specific purposes, such as simulating leaching conditions, predicting available concentrations, and long-term leaching (Izquierdo and Querol, 2012). However, most of these tests are single-step tests that may insufficiently predict the leaching potential of certain materials (Vamvuka and Kakaras, 2011).

The main factors that underlie the mobility of HMs must be explored to control the potential environmental hazards of using and disposing a by-product (Izquierdo and Querol, 2012). The chemical fraction of HMs is the most critical of these factors (Weiguo et al., 2009). Several extraction methods have been proposed in the literature to measure the chemical fraction in a material. Among these methods, the sequential extraction method of Tessier et al. (1979) is considered the best available method for assessing the origin, occurrence pattern, bioavailability, mobilization and transportation of HMs. The distribution of HMs among exchangeable, carbonate, oxide, organic and residual fractions can help to evaluate the mobility and ecotoxicity of HMs. Future studies must understand the leaching behavior and chemical speciation of HMs to facilitate the evaluation of the environmental risks and bioavailability of these metals. Accordingly, the leaching characteristics and chemical partitioning of ashes must be evaluated before their reuse (Grammelis et al., 2006).

In this paper, two co-firing (rice husk–sewage sludge and rice husk–dredged sludge blends) ashes were collected, and their basic characteristics (chemical composition, morphology and mineralogical composition) and environmental performance were examined via progressive toxicity characteristic leaching procedure and sequential chemical extraction. This study primarily aims to evaluate and compare the pollution levels and potential risks of trace elements (Cr, Cu, Fe, Mn, Ba and Zn) in the co-firing ashes. The basic characteristics and environmental properties of co-combustion ashes must be comprehensively investigated to establish an environmental and economical waste appraisal system.

2. Material and methods

2.1. Materials and ash samples preparation

The rice husk was collected from Wuhan, Hubei, and the sewage sludge samples were collected from a wastewater treatment plant in the same province. Dredged sludge samples were dredged from South Lake, which is seriously polluted by wastes from domestic and industrial sources. The raw materials were all placed under shade at room temperature during their transportation and storage until the drying procedure was completed. Afterward, the sludge (sewage and dredged sludge) and rice husk were dried in an oven at 105 °C for 3 d and 3 h to ensure complete dehydration.

China does not follow a relative standard for the co-combustion of sludge and rice husk. A typical incineration plant will burn sewage sludge in a fluidized bed furnace at 800 °C–900 °C (Donatello et al., 2010). Co-combustion below 1000 °C can effectively reduce the leachable content of HMs in ashes and conserve energy sources (Matsuzawa et al., 2006). The technology can also be applied in the existing incineration boilers. Using the GB/T 212-2008 method for the proximate analysis of coal, the ashes were prepared as follows. First, the muffle furnace with ventilation was heated at 815 ± 15 °C. Second, the sludge and rice husk mixture was placed in the furnace and co-combusted for 1 h. Third, the residue from the co-combustion was collected and grinded in a ball mill (XQM-4L) for 10 min. Fourth, the ashes were sieved through a 0.2 mm sieve.

High moisture content (≈80%) is a major deficiency in sludge treatment because a large amount of energy must be consumed for mass drying before combustion (Xiao et al., 2010). Compared with that consumed in the drying of raw sludge, the energy that is consumed in the drying of rice husk can be ignored in blended fuels. After eliminating the drying energy, the calorific value of blended fuels was calculated using Equations (1)–(3) as shown in Table 1 (Kliopova and Makarskienė, 2014).

$$Q_{\mathrm{drying}}^{\mathrm{theoretical}} = \frac{\mathrm{W} \times \left[\gamma + \left(\mathrm{T_{boiling}} - \mathrm{T_{room}}\right) \times \mathrm{C}\right]}{1 - W} \tag{1}$$

$$Q_{\mathrm{drying}}^{\mathrm{calculated}} = Q_{\mathrm{drying}}^{\mathrm{theoretical}} \Big/ \mathrm{E} \tag{2}$$

$$Q_{\mathrm{net}}^{\mathrm{as\ recieved}} = \omega_1 \times \mathrm{LHV_{Rice\ Husk}} + \omega_2 \times \mathrm{LHV_{sludge}} - \omega_2 \times Q_{\mathrm{drying}}^{\mathrm{calculated}} \tag{3}$$

where $Q_{drying}^{theoretical}$ and $Q_{drying}^{calculated}$ correspond to the theoretical and calculated drying energies in dry raw sludge, γ is the latent heat of water vaporization at 25 °C, C is the specific water heat (4.19 kJ/(kg °C), W is the moisture content (%), E is the drying efficiency (40%), ω_1 and ω_2 represent the rice husk and sludge content of the