Waste Management xxx (2018) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

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

Volatility and partitioning of Cd and Pb during sewage sludge thermal conversion

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ARTICLE INFO

Article history:
Received 27 August 2017
Revised 27 January 2018
Accepted 29 January 2018
Available online xxxx

Keywords:
Sewage sludge
Thermal conversion
Thermal analyses
Concentration ratio
Partitioning

ABSTRACT

In this paper, the thermal characteristics of sewage sludge and the transformation behavior of Pb and Cd during the thermal conversion process were addressed. The incineration process and pyrolysis process of the sewage sludge were investigated by thermogravimetric analysis. The results indicated that the thermal conversion process of the sewage sludge could be divided into three stages and the presence of oxygen could accelerate the decomposition of the sewage sludge. Furthermore, the effects of thermal conditions on the concentration ratio of Cd and Pb and their species partitioning in the residual char and ash were investigated. For the pyrolysis process, the maximum concentration ratio of Cd reached 41.64% at 500 °C and the lowest one 2.92% at 700 °C. Contrary, the concentration ratio of Pb remained above 93% as the temperature increased. Thus, the suitable temperature for the sewage sludge pyrolysis was below 500 °C. For the incineration process, the incineration temperature had great influence on the concentration ratio of Cd and Pb. When the incineration temperature increased from 700 °C to 900 °C, the concentration ratio of Cd decreased drastically from 99.32% to 10.96%. The maximum concentration ratio for Pb (95.31%) was reached at 800 °C. Besides, the lowest concentration ratio of Cd and Pb were obtained at a residence time of 30 min. The partitioning analyses of the Cd and Pb contained in the ash showed that the residence time had little effect on the partitioning of Cd and Pb, and the residual fractions of Cd and Pb were both above 90%. It was concluded that Cd and Pb were properly stabilized in the ash. Thus, Cd and Pb in the ash were difficult to be released into the environment and to cause secondary pollution.

1. Introduction

Sludge is a semi-solid residue produced from wastewater treatment, which contains biodegradable and recalcitrant organic compounds, as well as pathogens, heavy metals and other inorganic constituents. Among heavy metals, Cd and Pb are toxic for the human health especially the children's bone marrow, hematopoietic system and the nervous system (Magdziarz and Werle (2014); Taylor et al., 2014; Seggiani et al. (2012)). Therefore, the traditional sludge disposal methods, such as disposal in a landfill or the sea and reuse in agriculture, are subject to strict environmental restrictions. Nowadays, the disposal of sewage sludge is a growing concern throughout the world and in developing countries particularly (Kazi et al., 2005). At present, the thermal treatment which is a treatment method that yields a large reduction in volume, thermal destruction of toxic organics and recovery of the energy of organic sources and is acknowledged as the most efficient treatment method for sewage sludge (Lin et al., 2014). However, the emission of heavy metals during the thermal process is of paramount concern (Belevi and Moench, 2000; Song et al., 2004).

The thermal characteristics of sewage sludge and the influence of temperature and atmosphere have been investigated by thermal analyses (Otero et al., 2007; Viana et al., 2011; Varol et al., 2010; Wu et al., 2012; Magdziarz and Wilk, 2013; Cao et al., 2013). The studies on the pyrolysis, combustion and co-combustion of urban plant sewage sludge have been published by Calvo et al., (2004) and Ischia et al., (2007). Scott et al., (2006) investigated the kinetics of combustion processes by thermogravimetric analyses. Cao et al., (2014) and Wei et al., (2017) investigated the influence of atmospheres on the product distribution and the nitrogen transformations during the fast pyrolysis of sewage sludge.

The thermal behavior of the sludge composition is very complicated during the thermal conversion process. Therefore, it is still necessary to study the thermal behavior of the sewage sludge under different reaction conditions. Moreover, the previous studies have indicated that the partitioning of heavy metals among the

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https://doi.org/10.1016/j.wasman.2018.01.042
0956-053X/© 2018 Published by Elsevier Ltd.

incinerator discharges depends on the composition of sewage sludge, the operational conditions, the physical-chemical properties of the heavy metals, and the compounds formed during the combustion process (Wang et al., 2001). Thus, it is of paramount importance to study the effects of thermal conditions on the distributions and the bioavailability of Pb and Cd. Moreover, the bioavailability of Pb and Cd can be determined by the partitioning of the heavy metals in the ash (Chou et al., 2009).

In this work, the thermal behavior of the sewage sludge was investigated by thermogravimetric analysis and the influence of the incineration conditions on the concentration ratio and partitioning of Pb and Cd in the ash were also investigated using an electrically heated tube furnace.

2. Materials and methods

2.1. Sludge samples

12 identical sludge samples were collected from a sewage treatment plant in Hangzhou. The sludge samples were dried for 24 h at 105 °C in an electric oven, and then ground to pass through 110-mesh screen (particle size) by an agate mortar and pestle for the subsequent sample preparation. The dried sludge powders were stored in amber glass bottles. One of these samples was analysed for the contents of moisture, ash and volatile matter content by proximate analyses and for chemical composition by an Eurovector EA3000 CHNSO elemental analyzer (Singh et al. (2017)). The proximate and ultimate analyses of the sample are listed in Table 1 and heavy metal contents and ash analyses of the sample are presented in Table 2.

2.2. Experimental methods

2.2.1. Thermogravimetric experiment

The thermal characteristic experiments were carried out in a thermogravimetric analyzer (Q500, TA Instruments, America). For the thermal analyses (TG/DTG), the approximately 1 mg of the sample was placed in an alumina crucibles and heated from ambient temperature to 1000 °C at specific heating rate of 15 °C/min in 40 mL/min of air (incineration process) or argon flow (pyrolysis process). The atmosphere was also maintained during the heating and cooling process. The flow rate ensures the sample is in a suitable atmosphere during the run. Besides, the small volume of the sample and the slow heating rate ensure that the heat transfer limitations can be ignored. From these assays, the TG and DTG curves for each of the samples could be obtained as the outputs for the incineration and pyrolysis process.

2.2.2. Thermal experiment

The apparatus used in this experiment, as shown in Fig. 1, was composed of an electric-heated tube furnace, gas supplying unit and exhaust absorption device. The thermal processes of the samples were carried out in a quartz tube reactor with the body length of 1000 mm and the internal diameter of 36 mm. A quartz boat moved by a quartz rod was designed to feed the sludge sample into the reacting chamber. The temperature at the center inside of the reactor tube was monitored by a thermocouple and controlled by a programmed temperature controller. Moreover, the argon or air feed was regulated by a flow meter. In the downstream of the reactor chamber, a quartz-fiber filter was set up. During the thermal experiments, approximately 3 g of the sample was placed into a porcelain boat and pushed into the center of the quartz tube. The flow rate of air and nitrogen was 400 mL/min. For the pyrolysis run, the effects of the temperature (400–700 °C) on the concentration ratio of Pb and Cd in the residual char were investigated. For the combustion run, the effects of the temperature (700–900 °C) and retention time (10–60 min) on the concentration ratio and species partitioning of Pb and Cd in the residual ash were studied.

2.2.3. Chemical analyses

In this study, an atomic absorption spectrometer (AAS) (CONTRAA700, Jena, Germany) was used to test and analyze the contents of the target elements in the samples. To analyze the total heavy metal content, the samples (0.2 g) before and after heat treatment were digested in a microwave digestion device (WX-8000, Preekem, Shanghai) with acid mixture (HF, H_2O_2 and HNO_3). The concentrations of the target heavy metals in the digestion solution were analyzed using a flame atomic absorption spectrophotometer (AA-1800, Macylab, China).

An established multi-step sequential extraction method (Jameel et al., 2009) was used to extract the metals from the residual samples of about 0.5 g. The extraction was carried out in 50 mL polyethylene acid washed centrifuged tubes, which were also used for centrifugation to minimize the possible loss of solid. The following four fractions were collected during each extraction run and grouped according to the metal species: the acid

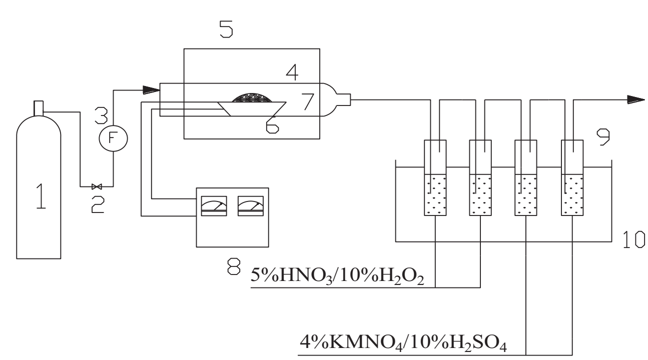

Fig. 1. Schematic of the tube furnace reactor system.

Table 1
Proximate and ultimate analyses of the sludge samples.

Proximate analyses w/%				Ultimate analyses w/%					$Q_{b,ad}$ /(J·g^{-1})
M_{ad}	A_{ad}	V_{ad}	$FC_{ad.diff.}$	C_{ad}	H_{ad}	N_{ad}	S_{ad}	$O_{ad.diff.}$	
2.37	45.81	45.47	6.35	24.63	3.32	2.96	1.06	19.85	11,141

Table 2
Heavy metal contents and ash analysis of the sludge.

Concentration of the heavy metals/(mg·kg^{-1})								Ash analysis/%							
Pb	Ni	Mn	Zn	Cr	Cu	Cd	As	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3
133.3	68.8	471.0	1840.0	167.0	1260.0	3.1	22.9	53.75	11.83	7.60	7.44	6.54	1.82	1.14	2.59

Please cite this article in press as: Zhang, Y.-f., et al. Volatility and partitioning of Cd and Pb during sewage sludge thermal conversion. Waste Management (2018), https://doi.org/10.1016/j.wasman.2018.01.042