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Influence of various chlorine additives on the partitioning of heavy metals during low-temperature two-stage fluidized bed incineration

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ABSTRACT

In this study, a pilot-scale low-temperature two-stage fluidized bed incinerator was evaluated for the control of heavy metal emissions using various chlorine (Cl) additives. Artificial waste containing heavy metals was selected to simulate municipal solid waste (MSW). Operating parameters considered included the first-stage combustion temperature, gas velocity, and different kinds of Cl additives. Results showed that the low-temperature two-stage fluidized bed reactor can be an effective system for the treatment of MSW because of its low NO_x, CO, HCl, and heavy metal emissions. The NO_x and HCl emissions could be decreased by 42% and 70%, respectively. Further, the results showed that heavy metal emissions were reduced by bed material adsorption and filtration in the second stage. Regarding the Cl addition, although the Cl addition would reduce the metal capture in the first-stage sand bed, but those emitted metals could be effectively captured by the filtration of second stage. No matter choose what kind of additive, metal emissions in the low-temperature two-stage system are still lower than in a traditional high-temperature one-stage system. The results also showed that metal emissions depend not only on the combustion temperature but also on the physicochemical properties of the different metal species.

1. Introduction

In order to solve problems arising from MSW, many developing countries are taking considerable interest in constructing MSW incinerators (Geng, 2012; State Council, 2012; Yu, 2012). However, this technology may give rise to considerable amounts of secondary products such as gaseous pollutants, fly ash, and bottom ash. Many studies have indicated that these byproducts may contain hazardous matters such as heavy metals and dioxins (Gupta et al., 2005; Jung et al., 2004; Vogg et al., 1987; Yasuhara and Katami, 2007). In Taiwan, incineration technology has been developed over 20 years. At present, there are 24 incinerators in Taiwan, and their annual treatment capacity is about 6.5 million tons MSW. However, these incinerators also produce about 290,000 tons of fly ash each year (TEPA, 2011). The Taiwan Environmental Protection Administration (EPA) has carried out long-term monitoring of these ash residuals. It has been found that heavy metals are a major concern. In traditional treatment processes, solidification/stabilization is the most common method for fly ash treatment. However, this costs lots of money and energy because of the additives used and the transportation. For the purpose of energy saving and carbon reduction, the current incineration procedure and its heavy metal control must be re-surveyed.

In general, MSW incineration employs air pollution control devices (APCDs) to control metal emissions. However, this practice is uneconomical because the equipment needs regular maintenance. In fact, the transportation of a heavy metal during waste combustion depends on its physical and chemical form (Corella and Toledo, 2000; Linak and Wendt, 1993; Nzihou and Stanmore, 2013; Yu et al., 2012). Volatile metals, which have higher saturated vapor pressures, enter the gas phase easily and re-condense on fine particles by homogeneous nucleation, heterogeneous condensation, coagulation, and agglomeration (Obernberger et al., 2006). For incineration with heavy metal emission control, Ho et al. (1993) have proposed two alternative technologies to improve the metal control efficiency. One is to use sorbents to capture the heavy metals during the incineration or after it. This method is efficient, but it is costly and requires secondary treatment of the waste sorbents. The other is to reduce the volatilization of metals by modification of the combustion operations. In 1995, the US government

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sponsored a plan to develop a low-temperature incineration system for treating radionuclide material (Wade and Williams, 1995). The results showed that emission of radionuclides and volatile heavy metals could be minimized. The advantage of this process is that it not only extends the lifetime of the APCDs, but also reduces the amount of fly ash and the secondary treatment cost. Therefore, the low-temperature incineration process is a potential technology to apply in waste treatment.

For the low-temperature incineration design, the unit should be composed of a primary combustion and a secondary combustion (afterburner). The fluidized bed incinerator (FBI) is an ideal reactor for low-temperature waste combustion because it can provide the ideal first combustion zone with good mixing, homogenized heat, and mass transfer (Ni et al., 2006; Suksankraisorn et al., 2010). In the previous study, Peng et al. (2014) developed a low-temperature two-stage fluidized bed combustion process for treating MSW. In the low-temperature combustion zone, the heavy metal emissions were efficiently controlled in the sand bed. Although a significant amount of unburnt carbon and CO were released through low-temperature combustion zone, but they were destroyed completely by the high-temperature secondary combustion zone.

In the fluidized bed incineration process, previous works have shown that the operating conditions may affect heavy metal control. Wey et al. (2001) pointed out the heavy metal emissions are related with the combustion temperature. Lin et al. (2010) noted that when the gas velocity decreases, it also reduces the turbulent mixing of the sand bed, causing poor heavy metal adsorption in the sand bed. Moreover, several studies have proved that the behavior of metal vaporization is highly influenced by the waste composition and reactive metallic species (Belevei and Monech, 2000; Yoo et al., 2002). Zhang et al. (2008) pointed out that sulfur can affect Cd emissions when it exists in different oxidizing and reducing environments. Wey et al. (1996) also found that organic and inorganic Cl additives may affect heavy metal emissions. Several studies have pointed out that chlorine reacts with metals to form low-boiling-point compounds (Chiang et al., 2001; Li et al., 2010). However, the effect of chlorine in this low-temperature two-stage fluidized bed incinerator on the control of heavy metals is not clear. Thus, this needs to be further investigated.

In this study, a low-temperature fluidized bed incineration process was carried out to evaluate heavy metal emissions using various Cl additives. An artificial waste mixture was selected, containing wood chips, polyethylene (PE), and heavy metals (Cr, Pb, and Cd). To confirm the pollutant control efficiency of the system, the gas pollutants of CO, NO_x, and HCl were monitored during the experiment. The following operating parameters were considered: (1) primary combustion temperature, (2) gas velocity, (3) organic Cl (polyvinyl chloride, PVC) waste feed, and (4) inorganic Cl (NaCl) waste feed. From the results, the efficiency of heavy metal control was estimated by the metal emissions and metal capture in different sand beds.

2. Materials and methods

2.1. Artificial wastes

Due to simulate municipal solid waste, the artificial waste was used in this study. It was composed of bald cypress wood chips (*Taxodium distichum*), polyethylene (PE) bags, and three kinds of nitrate metal solutions: $Pb(NO_3)_2$, $Cr(NO_3)_3{\bullet}9H_2O$, and $Cd(NO_3)_2{\bullet}4H_2O$. All the nitratre metals were extra pure reagent (purity > 99%) and supplied by Shimakyu. The wood chips and PE bag was obtained from Taiwan sawmill and commercially available product (Da-Ding company, China), respectively. The concentration of each metal was 0.5 wt.% as atoms of metals without nitrates. After the metals were dissolved in DI water, 1.3 mL of metal solution was added into the PE bags that contained the wood chips. For the different kinds of Cl additives, NaCl (inorganic Cl), and PVC (organic Cl) were selected. The extra pure reagent NaCl was supplied by Shimakyu. PVC was obtained from a commercially available rigid plastic raw material manufacturer (Formosa Plastics Corporation, Taiwan). After adjustment, the concentrations of both inorganic and organic Cl were 0.76 wt.% in the artificial waste. The feed rate of waste was 9.4 g/min. The detailed proximate and elemental analyses of materials are shown in the Supplementary materials.

2.2. Apparatus and experimental procedure

The experimental apparatus is a low-temperature two-stage fluidized bed system. This system was made by Jia-Ren heaters company, Taiwan. The schematic diagram of system and detailed experimental setup were described in a previous report (Peng et al., 2014). The fluidized media was silica sand which applied by Chih Chuen industry company in Taiwan. The density was 2600 kg/m^3. And the composition of sand was 97.8% SiO_2, 2.01% Al_2O_3, and 0.07% Fe_2O_3. In the first and secondary combustor, the silica sand was 330 g (545 μm) and 660 g (890 μm), respectively.

Before beginning the experiments, the first and second sand beds were set up at desired temperature. After reaching the specific temperature, the blower supplied a fixed flow air which controlled by the flow meter. Meanwhile, the initial gas composition was monitored by the multi-gas online analyzer (Horuba, PG-250, Japan) to confirm all tests were at the same condition. After temperature and gas composition became steady state, the experiments were performed. Each test was completed in 44 min. During the experiments, all fuel gases were cleaned by the scrubber tower system. For the bed material analysis, both sand beds were cooled down to room temperature and collected subsequently to analyze the heavy metals. Table 1 shows all the experimental condition in this study. During the experiments, flue gas and fly ash were sampled through isokinetic sampling by the Method 5 (USEPA, 1997). The detailed flue gas sampling setup were described in a previous report (Peng et al., 2014). For the metal analysis, all samples were extracted by microwave digestion using hydrofluoric acid (Extra pure reagent, Choneye) and nitrate (Extra pure reagent, Choneye). All the samples were analyzed by flame atomic absorption spectroscopy (FAAS) (Perkin AAnalyst 200, USA).

Hydrogen chloride (HCl) in flue gas was determined by TEPA (2008). First, particles in the flue gas were removed using a glass fiber filter. After that, HCl was absorbed by an impinger containing 100 ml 0.1 M NaOH (Extra pure reagent, Choneye). After the experiments, 1 mL 0.01 M mercury (II) thio-cyanate (Extra pure

Table 1
Operating conditions for each experiment.

Run	Temperature (°C)	Chlorine content (wt%)	Cl additives	
			PVC	NaCl
Run 1	800	0		
Run 2	800	0.76	●[a]	
Run 3	800	0.76		●
Run 4	550	0		
Run 5	650	0		
Run 6	750	0		
Run 7	550	0.76	●	
Run 8	550	0.76	●	
Run 9	550	0.76	●	
Run 10	550	0.76		●
Run 11	550	0.76		●
Run 12	550	0.76		●

[a] ●: The chloride additive was used in the test.