



Influence of ignition process on mineral phase transformation in municipal solid waste incineration (MSWI) fly ash: Implications for estimating loss-on-ignition (LOI)



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ABSTRACT

This research focused on the mineral phase transformation under varied ignition conditions with the objective of estimating loss-on-ignition (LOI) parameter in municipal solid waste incineration (MSWI) fly ash residues. LOI is commonly used to measure the volatile species, unburned carbon and moisture in the solid materials. There are criteria for LOI measurement in some research fields, while there is no standard protocol for LOI measurement in MSWI fly ash. Using thermogravimetry technique, the ignition condition candidates were proposed at 440/700/900 °C for 1 and 2 h. Based on X-ray diffractometry results, obvious mineral phase transformation occurred as a function of ignition temperature variation rather than ignition time. Until 440 °C, only some minor phases disappeared comparing with the original state. Significant mineral phase transformations of major phases (Ca- and Cl-based minerals) occurred between 440 and 700 °C. The mineral phase transformation and the occurrence of newly-formed phases were determined not only by the ignition condition but also by the content of the co-existing components. Mineral phase components rarely changed when ignition temperature rose from 700 to 900 °C. Consequently, in order to prevent critical damages to the original mineralogical composition of fly ash, the lowest ignition temperature (440 °C) for 2 h was suggested as an ideal measurement condition of LOI in MSWI fly ash.

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1. Introduction

Millions of tons of municipal solid waste are annually generated around the world. Incineration technology is deemed one of the commonest and most effective techniques for treating the generated waste in many countries, as it can reduce the waste volume by up to 90% and the waste mass by 70% (Nixon et al., 2013). As a renewable source of energy, this technology can also supply heat and electricity to the consumers (Lam et al., 2010). However, incineration comes with its own drawbacks, mainly associated with the cost, and the unavoidable generation of gaseous emissions (carbon dioxide and dioxins) and hazardous discharge (Orozco et al., 2007; Quina et al., 2010). Municipal solid waste incineration (MSWI) process produces a considerable amount of solid residues including bottom ash and fly ash that require proper treatments before disposal or recycling. Therefore, the studies about MSWI residues have raised concerns in the field

of waste management (Quina et al., 2008; Sabbas et al., 2003). Although the quantity of bottom ash is significantly larger than fly ash (Chimenos et al., 1999; Forteza et al., 2004), the latter is classified as a hazardous material due to the considerably higher concentration of leachable heavy metals and the other contaminants (Ferreira et al., 2003).

Various parameters have been employed to describe the physical, chemical and mechanical properties of the incineration residues (Ibáñez et al., 2000; Kim et al., 2010; Yang et al., 2014). Loss-on-ignition (LOI) is one of those parameters that refers to the mass loss of solid samples, which can be due to the loss of moisture, carbon, and so forth, when it is heated in an oxidizing atmosphere at specific temperatures. LOI is widely used to estimate the organic and/or carbonate content of different kinds of samples (Heiri et al., 2001). Ignition time and temperature were mentioned as the critical factors that can compromise the reproducibility and the comparability of the results (Hoogsteen et al., 2015; Konen et al., 2002). Various volatile salts, structural water and inorganic carbon may also contribute to LOI as a function of ignition temperatures (Sutherland, 1998; Dean, 1974). In addition,

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the ignition time may affect the existence of unstable components in the residues.

LOI has been employed for decades in several research fields as well as in the industry as a proxy for the characteristics of solid samples that requires varied ignition temperature and time for particular purposes. In the soil science, LOI is measured stepwise at 360–800 °C for 2–12 h for estimating the soil organic carbon, organic matter or inorganic carbon contents (Ghabbour et al., 2014; Massaccesi et al., 2015; Wang et al., 2013). In the field of geological science, the ignition conditions are controlled between 550 and 950 °C for over 2 h of exposure in order to estimate the organic and inorganic carbon contents (Santisteban et al., 2004; Wang et al., 2011). In the field of environmental science, LOI is used to provide a rough estimate of the amounts of total organic matter, volatile species (at 500/550 °C) and carbonates (at 900 °C) in the solid fraction of sludge when samples are heated for specific periods (Shathika Sulthana Begum et al., 2013; Vemic et al., 2015). LOI is also used in the field of combustion materials. In the coal-fired power generation industry, there has been a standard method of using LOI for measuring the carbon content of fly ash at the relatively high temperatures to describe the efficiency of the combustion process and to estimate further utilities (Burris et al., 2005; Styszko-Grochowiak et al., 2004; Zhang and Honaker, 2015). However, certain deviations, caused by the transformations of mineral phases during heating, were reported by some researchers (Brown and Dykstra, 1995; Vandenbergh et al., 2010). Particularly, LOI is an essential parameter that is commonly used for measuring the bulk chemical composition of the solid materials using X-ray fluorescence (XRF) technique (Husillos Rodríguez et al., 2013; Kiattikomol et al., 2001).

MSWI fly ash is sharing a certain resemblance with other combustion materials, for it is generated from the high-temperature combustion process, and is captured by an air pollution control (APC) system. In most cases lime is added to the system to remove the acid gases (Chen et al., 2012; Lin et al., 2003; Wang et al., 1998). However, MSWI fly ash has distinctive features in mineral and chemical compositions resulting from the complexity of raw materials, which may affect the proper condition of LOI measurement and the transformation of minerals during ignition. To date, no researches have specifically focused on this matter, and there has been no universal standard protocol for estimating LOI in MSWI fly ash until now. Besides, since a relatively precise LOI value is essential as a parameter in some laboratory analyses, the research on the influence of ignition process on the mass loss of MSWI fly ash and the mineral phase change is a prerequisite. The present research provides a fundamental reference to the influence of ignition process on mass loss and mineral phase transformation in fly ash with the prime objective to provide a directive for measuring LOI of fly ash residues under the optimized conditions. For this purpose, various ignition scenarios were designed and priority was given to the preservation of the original mineralogical composition. Efforts were made to prevent/minimize the breakdown of the original array of minerals, with special emphasis on the liberation of unbound moisture, the volatile species such as organic carbon, and the decomposition of unburned materials in the course of ignition process.

2. Materials and methods

2.1. Sample collection and preparation

The lime-treated MSWI fly ash samples were obtained from three anonymous incineration plants- S and R located in Fukuoka city, and K in Kurume city, Japan. These incineration facilities are used to treat the municipal solid waste generated by the corre-

sponding regions of the two cities. The facilities S, R, and K employ the stocker-type technology with the waste treatment capacity of 750, 900 and 300 tons per day, respectively. For the APC system, electrostatic precipitator is used in plant S, while fabric filters are used in plants R and K. The lime-treated fly ash samples used in this study were collected from the APC system. The original samples were well homogenized, freeze-dried, and then stored in air-tight containers before proceeding to the next step.

2.2. Thermogravimetry (TG)

Thermogravimetry (TG) was conducted to detect the relationship between the mass loss of the sample and the temperature in a TG-DTA 2000SA (Bruker AXS) apparatus. Approximately 20 mg of the freeze-dried fly ash sample was taken for each test and aluminum oxide (Al_2O_3) was used as the inert control material. Both the fly ash sample and aluminum oxide were simultaneously settled and heated in the air atmosphere and the tests were run in triplicate. The temperature program was set to elevate from the ambient temperature to 1100 °C in the step of 10 °C/min. The temperature was held constant at maximum for 15 min at the end of each test.

2.3. Ignition condition test

Fly ash consists of fine particles and a variety of mineral phases, which provide a property to easily absorb moisture from the atmosphere. To avoid the deviation caused by the moisture absorption, the stored fly ash samples were ground by mechanical milling machine for 1 min and then dried in an oven at 105 °C for over 6 h to attain a relatively constant mass before the ignition process. According to the curves obtained from TG tests, three conditions (440, 700 and 900 °C) were selected as the ignition temperatures. About 2 g of pulverized samples were placed in the capped crucibles, heated in a muffle furnace from the ambient temperature to the target temperatures, and then ignited for 1 or 2 h. The weights of the samples before and after ignition were recorded and the difference was noted as LOI through the following equation:

$$\text{LOI (\%)} = \frac{\text{mass loss of sample}}{\text{mass of original sample}} \times 100 = \frac{W_1 - W_2}{W_1 - W_0} \times 100$$

where W_0 , W_1 and W_2 are the masses of blank crucible, crucible with sample before and after ignition, respectively. The LOI measurements were launched in duplicate in each scenario, and the average LOI was deemed as the representative value.

2.4. Bulk compositional analysis

Major, minor, and, trace elements were determined by XRF (ZSX Primus II, Rigaku) using the pulverized samples described in Section 2.3. Considering that higher ignition temperature could induce structural change of the mineral phases by which extra components might contribute to the LOI results, the minimum LOI value obtained from 440 °C/2 h scenario was used as an essential parameter for conducting XRF analysis. Total carbon (TC) content was measured using the fly ash samples by TOC-Z (Shimadzu) combined with solid sample combustion unit (SSM-5000A, Shimadzu) at 900 °C.

2.5. Mineral phase detection

The mineral phases of the samples before and after ignition in each condition were analyzed by X-ray diffractometry (XRD, Rigaku Multiflex) using $\text{Cu K}\alpha$ radiation generated at the voltage

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