Emission Characteristics of PM$_{10}$ during Sewage Sludge Combustion

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ABSTRACT

Combustion of sewage sludge was conducted in a laboratory-scale drop tube furnace to investigate the emission characteristics of PM$_{10}$ (particles with the aerodynamic diameter smaller than 10 $\mu$m). The experiment temperature was varied from 1173 to 1573 K. The results show that the emissions of submicron particulate matters (PM$_1$, particles with the aerodynamic diameter smaller than 1 $\mu$m) and supermicron particulate matters (PM$_{1-10}$, particles with aerodynamic diameter ranging from 1 $\mu$m to 10 $\mu$m) both decrease with temperature increasing. Meanwhile, the morphology of PM$_{10}$ was also analyzed by X-ray fluorescence (XRF) and scanning electron microscopy (SEM) to further understand its formation mechanisms. The results indicate that PM$_1$ is mainly formed from the vaporization and condensation process and its major elemental compositions are Na, S and P, while the agglomeration of melting minerals and the fragmentation of minerals contribute to PM$_{1-10}$, which is chiefly composed of Si, Al and Ca.

Keywords: Sewage sludge; Combustion; PM$_{10}$.

INTRODUCTION

Sewage sludge is the mainly byproduct of wastewater treatment. It was reported that the sewage sludge production of China in 2010 was about 18.00 million tons, and the annual increase rate was 8–10% (Ma et al., 2010). As it contains abundant organics, heavy metals and disease-causing micro-organisms, how to dispose sewage sludge has become a serious environmental issue. At present, the primary disposal methods include agricultural utilization, landfill and incineration. However, agricultural utilization and landfill have been gradually restricted as a result of their inherent disadvantages. Incineration has been widely applied in sewage sludge disposal due to its unique characteristics like volume reduction, organic micro-organisms destruction and energy recovery. For example, the fraction of the sewage sludge disposed by incineration in Japan and Europe were 70% and 30%, respectively (Yao and Naruse, 2009). However, a large amount of NO$_x$, SO$_2$, heavy metals and particulate matters were produced during sewage sludge combustion. It is well known that particulate matters have an important impact on environment and human (Braniš and Větvička, 2010; Kim et al., 2010; Lee et al., 2011), especially PM$_{10}$ is enriched with heavy metal and acids as a result of its abundant surfaces. Furthermore, PM$_{2.5}$ cannot be captured effectively by the current air pollutants control devices (APCDs) (Yao et al., 2010). These escaped PM$_{2.5}$ can stay atmosphere for a long time and have a possibility to be inhaled into the lung. Hence, the research about PM$_{10}$ (Begum et al., 2010; Avino et al., 2011; Colbeck et al., 2011), especially from the combustion source has been a hot wave in the world (Wand et al., 2009; Yao et al., 2010; Calvo et al., 2011).

Over the past few decades, the emissions of PM$_{10}$ and its formation mechanisms have been studied extensively. It was believed that the vaporization-nucleation-condensation process of the inorganic matter contributed to the submicron particulates (PM$_1$) (Quann et al., 1982), which was dominated by the elements of S, Si, P, and Na (Zhang et al., 2006). While the supermicron particulates (PM$_{1-10}$) was considered to be formed from the coalescence of melted minerals and the fragmentation of coke and mineral particles (Miller and Schobert, 1993), and the literatures suggested that the main constituents in PM$_{1-10}$ were Ca, Fe, Al, and Si (Wang et al., 2008). In addition, the influences of various parameters were considered, including the coal type (Takuwa et al., 2006), coal particle size distribution (Ninomiya et al., 2004), combustion temperature (Buhre et al., 2006; Liu et al., 2007), oxygen content of combustion (Sui, 2006), alkali,
sulfur content in coal (Buhre et al., 2005), the addition of limestone or kaolin (Zhang et al., 2002; Ninomiya et al., 2009; Chen et al., 2011), the activities of Ca-based, Mg-based and Fe-based species (Ninomiya et al., 2010), coal blending (Buhre et al., 2006) and co-combustion of sewage sludge and coal (Wang et al., 2009).

The previous studies of properties of particulate matters (PM) were mainly about coal combustion or co-combustion of coal and sewage sludge. Compared to coals, the volatile matter and ash is very high in sewage sludge. Meanwhile, there is not organic mineral matter, which can affect the formation mechanisms of particles during combustion process (Werther and Ogada, 1999). Hence, the emission characteristics of particles during sewage sludge and coal combustion are different. The aim of this study is to reveal the emission characteristics of PM$_{10}$ during sewage sludge combustion in a laboratory scale drop tube furnace.

**METHODS**

The sewage sludge used in this experiment was sampled from a wastewater treatment plant in Wuhan, China, which was a typical municipal sludge. Prior to use, sewage sludge was dried, milled and sieved to less than 74 μm. The proximate and ultimate analysis of sewage sludge was summarized in Table 1, and the composition of ash was shown in Table 2.

The experiments were conducted in a laboratory-scale drop tube furnace (DTF) with a 56 mm internal diameter and a 2000 mm vertical length. The DTF consists of a feeding system, a high temperature furnace and a sampling system, as shown in Fig. 1. The raw materials entrained in the primary airflow were fed into the DTF by a micro feeder (Model MFEV-10, Sankyo Piotech Co. Japan), and the feeding rate was 0.3 g/min. The furnace is electrically heated by three sections and the temperature of the furnace was automatically controlled by thermocouples. The experimental temperatures were 1173, 1373 and 1573 K. The primary airflow was mixed with the feed fuels prior to enter the furnace, and the primary airflow feed rate was 1 l/min. The secondary airflow feed rate was 4 l/min. The sample residence time in the furnace was about 2 s in all runs. The sampling system is consisted of an isokinetic sampling pipe, a Dekati cyclone separator (Model SAC-.65), a Dekati low pressure impactor (DLPI) and a vacuum pump. To minimize the oxidation as well as particle-particle

| Table 1. Ultimate and proximate analysis of sewage sludge (W%). |
|---------------------------------------------------------------|
| Ultimate analysis/% (daf) | Proximate analysis/% (ad) |
|---------------------------|---------------------------|
| C  | H  | O (diff) | N  | S  | A  | M  | V  | FC |
| 22.91 | 4.06 | 68.20 | 4.04 | 0.79 | 45.53 | 2.25 | 45.39 | 6.83 |

| Table 2. Composition of ash in sewage sludge (W%). |
|---------------------------------------------------|
| Si | Al | Fe | Ca | Mg | K | Na | Ti | S | P | Mn | Cu | Zn |
| 51.61 | 16.65 | 7.39 | 3.66 | 3.89 | 2.95 | 0.66 | 0.70 | 2.37 | 9.87 | 0.12 | 0.03 | 0.1 |

![Fig. 1. The schematic diagram of drop tube furnace (DTF).](image-url)
interaction, the exit gas entraining particles was initially quenched and diluted with N₂ and collected by a water-cooling sampling probe. Subsequently, the particulate matters more than 10 μm were collected by the cyclone separator, and the remainders, i.e. submicron and supermicron particles, were directed to the DLPI for size-segregated collection, and the DLPI used here is composed of 13 stages having the aerodynamic cut-off diameters ranging from 0.0281 to 9.8 μm. The elemental composition and morphology of the collected particles were analyzed by XRF (Mode EAGLE III, EDAX Co, America) and SEM/EDX (Mode Sirion-2000, FEI Co. America), respectively.

RESULTS AND DISCUSSION

Influence of Temperature on PM₁₀ Emissions

The emission of PM₁₀ during sewage sludge combustion as function of temperature was shown in Fig. 2. The emissions of PM₁ and PM₁–10 decreased with temperature increasing from 1173 K to 1573 K, which is contrary to that of coal combustion (Zhang et al., 2006; Liu et al., 2007). This phenomenon may be related to the different mineral compositions between sewage sludge and coal. Although high temperature has a positive effect in the formation of submicron particles due to more minerals vaporization during sewage sludge combustion, the interactions between the vaporized minerals and refractory minerals were also significantly enhanced, which results in the reduction of PM₁₀. Additionally, it is also found that the PM₁₀ distribution presents “V” shape between 0.1 and 4.4 μm, and there is a peak at 0.1 and 4.4 μm, respectively, which is similar to the PM₁₀ emission during pulverized coal combustion studied by Sui (2006).

Elemental Analysis of PM₁₀

To further understand the PM₁₀ characteristics, the particles with different size were analyzed by X-ray fluorescence (XRF) for elemental composition qualification. The elements detected in experiment include Na, K, Mg, Si, Al, Ca, S, P, Fe, Mn and Ti. Fig. 3 demonstrates the elemental compositions of PM₁₀, PM₁ and PM₁–10.

Fig. 3(a) indicates that PM₁₀ mainly consists of Na, Mg, Si, Al, Ca, S and P especially the contents of Na, Si, Al, S and P all are more than 5%, whereas less than 2% Fe, Mn and Ti were found in PM₁₀. Additionally, both of the percentages of Na and Mg in PM₁₀ have an evident increment with the temperature rising from 1173 to 1573 K, while Si and Al decrease significantly, P and Fe have a tendency to decrease and K, Mn and Ti hardly change.

The distribution of Na, S, P, Si and Ca in PM₁ is given in Fig. 3(b), and the content of Na and S is far more than that of Si and Ca. At the same time, the melting and boiling point temperature of Na and S is lower than those of Si and Ca. Hence, it can be concluded that PM₁ produced during sewage sludge combustion is mainly formed from the vaporization and condensation of volatile elements. Additionally, the contents of Na and S with low volatile temperature don’t have an interrelation to the temperature variation. However, the refractory elements such as Si, Al, Ca and Fe in PM₁ are increasing when the temperature increases.

The major components of PM₁–10 are Si, Al, Ca, Na, Mg and P, which is shown in Fig. 3(c). As the temperature rises from 1173 to 1573 K, the contents of Na, Mg and S all are increased, so that the interaction between the volatile element vapors and refractory minerals are enhanced significantly with the increment of temperature, which leads to the formation of coarser particles. On the contrary, Si, Al, Ca, P and Fe are decreased when temperature increases, especially Si, P and Al decrease markedly, which
is the major cause of the reduction of PM\textsubscript{1-10} with the temperature increasing. Si and Al with high melting/boiling point mainly exist in PM\textsubscript{1-10} due to minerals agglomeration or fragmentation during combustion. However, the elements with low melting point such as Na and S are primarily distributed in PM\textsubscript{1} and the fractions of Na and S in PM\textsubscript{1} are more than 15%.

**Morphology Analysis of PM\textsubscript{10}**

The typical morphology of the fourth stage particles (0.154 μm in diameter) collected by DLPI at 1373 and 1573 K are demonstrated in Fig. 4. It is obviously observed that the particles produced at 1373 K is mainly in the form of cluster or chain agglomerate structure, while the typical morphology under 1573 K is primarily irregular fractal agglomerate structure. The results indicate that the combustion temperature has an important influence on submicron particulate formation.

Fig. 5 shows the typical morphology of the tenth stage particles (2.36 μm in diameter) collected by DLPI at 1373 and 1573 K. It was found that most of them are sphere with smooth surface, and some are sphere-liked with smooth surface. The smooth surface of particles indicates the occurrence of minerals melting during combustion, which further proves that the agglomeration of melting minerals contributes to the formation of PM\textsubscript{1-10}. Additionally, there are also some particles formed by adhering of some irregular and sphere particles.
CONCLUSIONS

Temperature has an important influence on particulate emission during sewage sludge combustion. The enhanced interactions between the volatile and refractory minerals with temperature increasing lead to the formation of coarser particles and the reduction of PM$_1$ and PM$_{1.10}$.

PM$_1$ is mainly formed from the vaporization and condensation of volatile elements including Na, S and P, while the agglomeration and fragmentation of minerals contribute to PM$_{1.10}$ formation. PM$_{1.10}$ is chiefly composed of the refractory elements including Si, Al and Ca. The morphology of PM$_1$ is mainly irregular agglomerate structure including cluster, chain and fractal structure, while most of PM$_{1.10}$ are sphere with smooth surface.

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