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Migration and transformation of sulfur in the municipal sewage sludge during disposal in cement kiln

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ABSTRACT

The aim of this work was to investigate the migration and transformation of sulfur in the municipal sewage sludge during disposal in cement kiln, and better understand the emission of the sulfur related pollutants in this process. In consideration of the temperature conditions in the practical operation, municipal sewage sludge was pre-dried at 105 °C, and then dried at 210, 260 and 310 °C, co-combusted with cement raw mill at 800, 900 and 1000 °C, and 1350, 1400 and 1450 °C respectively in the laboratory. X-ray photoelectron spectroscopy (XPS) was used to determine the S2p spectral lines of the municipal sewage sludge treated in the different process. Besides, The Thermal Analysis-Thermogravimetry (DTA-TG), Back Scattered Electron (BSE) and Energy Dispersive Spectrometer (EDS) were also employed to explore the mechanism of sulfur subsistence at 1450 °C. The results indicate that sulfide, thiophene, sulfone and sulfate are mainly sulfur compound in the municipal sewage sludge dried at 105 °C. Sulfoxide, a new sulfur compound, appears after it is further dried at 210 °C. The relative contents of sulfide and thiophene are continuously declined as the drying temperature increases due to their evaporation, decomposition and transformation in this process. The transformation of sulfide and thiophene makes the relative contents of sulfoxide and sulfate accordingly increased. However, the relative content of sulfone experiences an elevating-lowering process while the dry temperature elevated from 210 to 310 °C. This case is related to its evaporation and decomposition, as well as its production for the transformation of sulfide and thiophene. In the co-combustion process, sulfide, thiophene and sulfone are entirely vanished for their evaporation, decomposition and transformation. Sulfone is still contained at 800 °C, but when the temperature unceasingly rises, it is completely decomposed or evaporated and sulfate is the only sulfur compound. The microstructures left by the gas release are also observed in the mixtures sintered at 1450 °C, however sulfate still exists even at 1450 °C. The BSE and EDS results show that the melt phase is the important contribution to the appearance of sulfate at the high temperature. These results will sever as a theoretically reference for the pollution control of the sulfur related pollutants in the disposal process of the municipal sewage sludge in cement kiln.

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1. Introduction

With the increasing population, industrialization, and effluent quality standards, sewage sludge is largely generated from municipal waste water treatment worldwide and its production keeps growing (Qin et al., 2015; Font et al., 2011; Fernandez-Anez et al., 2014). Currently, the treatment of municipal sewage sludge attracts ever-increasing importance in our society for the fact that the people's living standard and their environmental awareness have been improved (Mills et al., 2014). In addition, the municipal sewage sludge has threaten public health, soil resources and water resources, also has made an emission to the atmosphere resulting air pollution (Davids et al., 2017; Caraballo et al., 2017). The traditional methods for the treatment of municipal sewage sludge are mainly the landfill (Yang et al., 2017), agriculture (Tervahauta et al., 2014) and incineration (Xu et al., 2017). Landfill has been getting more popular because of its low investment, large capacity in the treatment rate which is relatively low in the vast number of developing countries, but the landfill leachates pollutant matters, and also the sludge composition is complex, once the impermeable infrastructure facilities do not run well, these pollutant matters will enter the underground water and contaminate groundwater, and then the secondary pollution will be resulted to the more useful land and water resources (Alibardi and Cossu, 2016). The utilization of the municipal sewage sludge in the agricultural means

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promoting the agricultural production by using municipal sludge digestion, as composting or compound fertilizer to provide the nutrients and some trace elements for plant growth. However, the heavy metals in the sludge will accumulate continuously in the food and enter the human body via the food chain in the end (Krzyzanowski et al., 2016). The incineration as a more thorough method for the sludge disposal, can eliminate the toxic and harmful organic matters, also recycle the energy. But it generates harmful substances in the incineration process and then causes a secondary pollution (Zhao et al., 2017). More importantly, the incineration ash has to be subsequently disposed (Li et al., 2012). In short, the traditional approaches to treat the municipal sewage sludge have various limitations (Zhang et al., 2013). It was reported that using cement kiln for municipal sewage sludge management has many advantages compared with other methods (Xu et al., 2014). These advantages can be summarized as (Rovira et al., 2010, 2011): (i) high temperature in kiln results in the avoiding of the toxic organic materials and the bacteria; (ii) most heavy metals can be immobilized into the cement clinker; (iii) fossil fuels can be reduced due to the high calorific value of sludge (Trezza and Scian, 2000; Dewil et al., 2006). In addition, as municipal sewage sludge itself contains SiO_2, CaO, Al_2O_3, Fe_3O_4 and other components of cement (Zabaniotou and Theofilou, 2008; Valderrama et al., 2013), it can be also used as an alternative raw material (Lin et al., 2012; Yan et al., 2014). Thus, this method has received extensive attention. Xu et al. (2014) have investigated the feasibility of using sludge as resource for producing cement clinker. Besides, the burnability of raw mill, the formation, morphology and crystalline composition of the clinker, and the strength of cement are all discussed in their work. It is noteworthy that municipal sewage sludge contains the nitrogen- containing compounds, organic matters and heavy metals (Chen et al., 2012), thus the NO_x, NH_3 and PAHs emissions were also studied (Lv et al., 2016). In addition, there also contains various sulfur compounds and their contents are high. Then, the emissions of air pollutants related to sulfur are complicated during the disposal in cement kiln (Li et al., 2009). Therefore, the studies on migration and transformation of sulfur will help people better understand the emission of sulfur related harmful pollutant in the municipal sewage sludge during this disposal. Various sulfur compounds in the municipal sewage sludge have reported by the researchers (Dote et al., 1992; Harrison et al., 2006; Kienhuis and Geerdink, 2000; Merino et al., 2007), and also the sulfur forms and transformation during the co-combustion of sewage sludge and coal were investigated by Li et al. (2009). Besides, the solidification of the sulfur based hazardous waste was studied (Mohamed and El Gamal, 2007), the emission, mass balance and distribution characteristics of PCDD/Fs and heavy metals during co-combustion of sewage sludge and coal in power plants were accessed by Zhang et al. (2013), cement kiln dust was solidified using sulfur binder (Mohamed and El Gamal, 2011), the sulfur dioxide oxidation, cycle assessment and release in the cement kiln also were investigated (Caraballo et al., 2017; Dellinger et al., 1980; Nielsen et al., 2011).

Although there is a lot of research efforts as reported in these literature, it is difficult, through them, to correctly determine the sulfur forms and understand their migration and transformation during disposal in cement kiln because of the different component, technology system and temperature condition. There still remain some important points needed to be systematically clarified. These points are important to understand the emission of the sulfur related pollutant during this process. Therefore, the municipal sewage sludge was collected and then it was treated under various conditions. The S2p spectral lines were tested by XPS, and then the migration and transformation of sulfur during this process were discussed. Besides, the mechanisms of the sulfur migration and transformation during this process are also explored in this work.

2. Experimental

2.1. 1Materials

The municipal sewage sludge used in this study was collected from Shanghai Quyang Sewage Treatment Plant. It was fresh sludge containing more than 80% water after the initial mechanical dewatering. $CaCO_3$, Al_2O_3, SiO_2 and Fe_2O_3 all are chemical reagents, and they were used to prepare the cement raw material. The chemical components of the municipal sewage sludge dried at the 105 °C to the constant weight are given in Table 1.

2.2. Sample preparation

The fresh sludge was collected and it was pre-dried at the 105 °C to the constant weight. Then the pre-dried sludge was acquired. According to the current disposal technology of municipal sewage sludge in cement kiln, three drying temperatures such as 210, 260 and 310 °C were selected to simulate the temperature condition of the residual heat from the cement kiln inlet, and then the pre-dried sludge was ground into about 0.08 mm size and further dried at these temperatures to the constant weight. After further drying, the dried sludge with the residual heat from the cement kiln inlet was prepared, and it was ground into about 0.08 mm size. 20% dried sludge with the residual heat from the cement kiln was mixed into the cement raw mill. The chemical components and the parameters of cement raw mill are given in Table 2. The mixture of the sludge dried at 260 °C and cement raw mill were co-combusted at 800, 900 and 1000 °C in the cement precalciner kiln for 1 h. After this process, this mixture treated at 1000 °C was continuously sintered at the temperatures of 1350, 1400 and 1450 °C in the cement rotary kiln for 1 h. The detailed disposal process and corresponding treated temperature are showed in Fig. 1.

2.3. Methods

X-ray photoelectron spectroscopy (XPS) was used to analyze the sulfur compounds, relative content and forms of the municipal sewage sludge after treating. The ESCALAB 250Xi from American Thermoelectric Company was used, and the analysis chamber vacuum degree was 4.3×10^{-10} mbar, the energy resolution Ag3d5/2, FWHM0.44 eV, the sensitivity 3.28 Mcps. The area of peak reflected the relative content, and the height contained information strength in S2p spectral line.

In order to observe the thermal process of calcium sulfate, Differential Thermal Analysis-Thermogravimetry (DTA-TG) method was used by NETZSCH-STA 449C thermal analyzer with a heating rate of 10 °C per min. The atmosphere is oxygen-free nitrogen.

The mixture sintered at 1450 °C was put into the air dryer. It was retained to observe the microstructure with a Quanta 200 FEG field emission environmental scanning electron microscope (SEM). The accelerating voltage was 20 kV, and the magnification was adjusted to 2000.

Further sample preparation is necessary for the Back scattered Electron (BSE) image acquisition. The dried specimen was vacuum impregnated with low viscosity epoxy resin. After epoxy drying, it was ground with SiC paper of 320, 500, 800, 1200, and 2400 grit for about 4 min each. Subsequently, it was polished with diamond paste of 3, 1, and 0.25 μm for around 2 min each and cleaned up with low-relief polishing cloth. The polished sections used for BSE imaging were further investigated with energy dispersive spectrometer (EDS) analysis. In this work, EDS region analysis was performed on the mixture co-combusted at 1450 °C. The accelerating voltage was chosen at 20 kV.

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