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Distributional and compositional insight into the polluting materials during sludge combustion: Roles of ash

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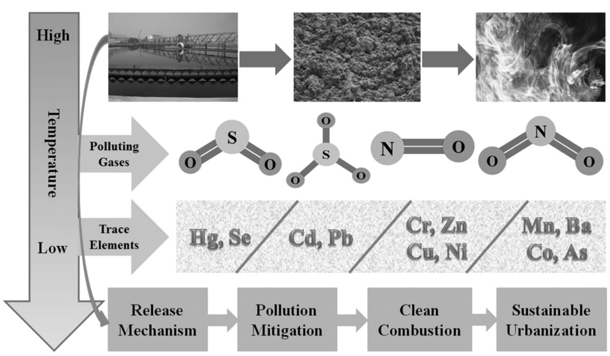
GRAPHICAL ABSTRACT

In this study we systemically clarified the mineral phases and distributions of trace elements during sludge combustion as well as the polluting gases and identified the roles of sludge ash for the first time. Accordingly, we can establish a foundation for controlling and mitigating the environmental impacts toward clean combustion and even the sustainable urbanization in China.



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ABSTRACT

Here we systemically elucidated the compositions and distributions of polluting materials during sludge combustion using both experimental and theoretical methods where the roles of ash were clarified in a category within aluminosilicate system. With regard to the polluting gases, an increasing temperature increased the SO_2 release in replace of SO_3 and sulfates while continuously enhanced the formation of NO and NO_2. Comparatively, an increasing input oxygen amount had limited effect on S-bearing gases whilst continuously increased the NO_x release. Additionally, the ash had a sulfur fixation effect through inducing the transformation from oxides to sulfates. The trace elements were overall categorized into four types based on their volatilities and the distributions and mineral phases of them were then clarified in detail. It was proved that with increasing temperature, they were increasingly distributed in gaseous state with ash providing a fixation effect. Moreover, 5 kinds of decomposition reactions related to polluting materials, namely sulfates, oxides, arsenates, silicates and hydroxides, were deeply analyzed from a respect of Gibbs energy where their stabilities and roles of ash were identified. Furthermore, the sludge combustion was experimentally clarified using a TG-MS system and here we defined a new parameter, namely peak index, to characterize the releases of polluting gases. This not only deepened the understanding of sludge combustion experimentally but also partially validated the theoretical

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analysis in quantity. Most importantly, the present study contributed to establishing a foundation for controlling and mitigating the environmental impacts of polluting materials toward efficient and clean sludge combustions.

1. Introduction

Nowadays China is promoting urbanization and poverty eradication where the proportion of people dwelling in cities has risen from ∼18% in 1978 to more than 55% in 2016 [1,2]. The expanding urbanization not only results in the improvement of living standard but also brings up the increasing challenges to maintain the urban environment. Amongst them the requirement of wastewater treatment has greatly grown in urban areas. In 2014, the emission of sewage in China has been increased to 71.6 billion tons and this generated huge production of sewage sludge, up to 28 million tons [3]. This sewage sludge, after dewatering and drying, needs to be treated timely and appropriately with the avoidant of secondary pollutions.

Many routes have been developed to dispose the sewage sludge, such as anaerobic digestion, combustion as well as thermal-chemical methods including gasification and pyrolysis. Due to the simple engineering operations, large treatment capacity and low costs, the sludge treated by combustion or incineration remained more than 20% [4], accounting for a key method for sludge treatment. During the sludge combustion, control and mitigation of polluting materials, including polluting gases and toxic trace elements, makes up the crucial problems to be addressed. In these contexts, the clarification of release mechanism of polluting gases and trace elements is of great meanings toward an efficient and clean combustion.

However, there are quite limited researches systemically investigating the thermodynamic distributions of polluting gases and trace elements during sludge combustion. To bridge this gap, the present study was motivated, which accounted for the first innovation. Another significant issue related with sludge combustion is the effect of sludge ash, which has been barely discussed previously. Generally, the sewage sludge has high content of sludge ash and the ash are abundant in SiO_2 and Al_2O_3 [5–7], i.e., it can be summarized and included in a typical aluminosilicate system. In terms of chemical equilibrium, the presence of sludge ash could remarkably influence the releases of polluting gases and trace elements. Additionally, identifying effect of sludge ash on the combustion results provided other significant meanings. Under a flowing agent, the combustion process is more analogous to the phenomenon in absence of sludge ash since the volatiles and the released materials would be rapidly taken away. However, under a fixed agent, it is more similar to that in presence of sludge ash since the contacting and reaction time between the released materials and sludge ash is enough. Thus the influence of sludge ash and agent amounts on the combustion results would be systemically analyzed in this study, which accounted for another significant innovation here.

In particular, for the trace elements, in addition to distributions, another scientific issue is the transformation mechanism of mineral phases during sludge combustion. Generally with varying combustion temperature, the equilibrium mineral phases containing trace elements would get changed. Especially in presence of sludge ash, in theory, the releases of trace elements could be analyzed in the category of aluminosilicate system. Moreover, all the experimental results in previous studies related to trace elements releases during sludge combustion could be discussed and summarized in this category. In other words, we can establish a foundation for the transformation mechanism analysis of these mineral phases containing trace elements in a quite new respect, which accounted for another key innovation here. Accordingly, the clues for control and modification of trace elements could be further identified toward clean sludge combustion and even the sustainable urbanization.

2. Material and methods

2.1. Characteristics of raw materials

In order to analyze the releases of polluting gases and trace elements during sludge combustion, the first step necessary should be determining the chemical compositions of raw sludge. Since the sludge collected from different places could be quite variable, a typical and representative one should be selected appropriately. With respect to that, not only a series of previous studies [8–12] were referred to but also an experimental measurement of raw sludge was performed, collected from Nanshan Wastewater Treatment Plant in Shenzhen, China. The results of proximate and elemental analyses are listed in Table 1 and herein not only the major elements of C, H and O but also the minor ones of N and S were considered. Accordingly the hypothetical molecular formula of organic materials in sludge could be deduced as $CH_2O_{0.65}N_{0.1143}S_{0.025}$ and in order to simplify the calculations, it was assumed that only one mole of sludge was input the combustion system.

Since a main innovation of the present study was analyzing the effect of sludge ash, herein the experimental analysis of sludge ash after combustion was conducted using the X-ray fluorescence (XRF, S4-Explorer, Bruker) technique. In addition, a series of studies were also refereed to [13–16]. The results are shown in Table 2 and it can be observed that, the components with high contents were SiO_2 and Al_2O_3, typical network forming oxides in silicate system, whereas the minorities with contents less than 5% were Na_2O and K_2O, typical network breaking oxides.

With regard to the trace elements, their concentrations could cover a quite large scale for different types of sludge and similarly, both literature survey and experimental measurements were conducted [17–20]. The concentrations of these trace elements are listed in Table 2 and herein 12 kinds of trace elements were taken into account, including Zn, Mn, Ba, Cu, Cr with the concentrations higher than 100 ppm, Pb, Ni with the concentrations higher than 10 ppm and As, Co, Se, Cd, Hg with the concentrations less than 10 ppm.

2.2. Principles of thermodynamic calculations

After determining the chemical compositions of raw materials, the theoretical releases of polluting gases and trace elements could be identified using the FactSage software [21] and before that, two issues should be declared firstly. One is determination of the amounts of air input the combustion system and the other one is clarification of the principles of thermodynamic calculations. With regard to the former one, the reaction between sludge and oxygen could be described as Eq. (1). Thus by means of Eq. (1), it was calculated that at least 1.3143 mol of oxygen was required for the complete combustion of 1 mol of sludge in the temperature range of 600–1200 °C. In addition to temperature, the amount of oxygen input was taken as another variable considered here and consequently, we can obtain a series of 3D colormaps. With

Table 1
Proximate and elemental analysis of the sludge (dry basis, wt%).

Proximate analysis		Elemental analysis	
Moisture	5	C	30
Volatile	50	H	5
Ash	33	O	26
Fixed Carbon	12	N	4
		S	2