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Environmental mitigation of sludge combustion via two opposite modifying strategies: Kinetics and stabilization effect



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GRAPHICAL ABSTRACT

In this study we deepened the understanding of distribution behaviors of hazardous elements including trace elements and polluting gases during sludge combustion. The acidic kaolin and alkaline CaO additives were used for pollutant stabilizations, representing two general strategies. The additives themselves underwent great phase changes as the temperature increases, so there are different mechanisms of reaction and adsorption about trace elements and polluting gases in different temperature ranges.



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ABSTRACT

Nowadays, environmental mitigation remains a key issue challenging sludge combustion toward its clean utilization. Herein, two opposite additives, CaO and kaolin, representing two different strategies, were employed for environmental mitigation of sludge combustion. An advanced TG-MS (thermogravimetric-mass spectrometer) system was used. The online MS analysis indicated that kaolin remarkably decreased the release of NO_x whereas the fixation effect of CaO on SO_2 was more pronounced. Furthermore, the experiments using a fixed tube furnace proved that overall kaolin had a better role controlling volatility of trace elements (Cr, Ni, Cu, Zn, Pb, As and Ba) than CaO and we found that the fixation mechanism of CaO changed at 750 °C, whereas that of kaolin changed at 500 °C. Then the kinetics of the various stages during sludge combustion were investigated. We found that low temperature stages were along with the formation of new phases, whereas, at high temperatures, the diffusion could be rate-controlling step. The present study thus provides not only a deep understanding of sludge combustion mechanisms but also two effective modifying strategies toward the environmental mitigation of sludge combustion.

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1. Introduction

With the acceleration of urbanization, more and more municipal sewage treatment plants are built and as a result, the production of sewage sludge is increasing rapidly. Indah Water Konsortium Sdn. Bhd. recorded that in the year of 2020 there will be about 7 million cubic of sewage sludge produced annually [1]. As a by-product, sewage sludge is a complex and heterogeneous material composed of microorganisms, heavy metals, inorganic minerals and organic toxicants [2–4], which are harmful to human being and the surrounding environment. In another perspective, the dehydrated sludge can be taken as renewable fuel because it is rich in organics with high calorific values [5]. Particularly, the global warming necessitates the utilization of renewable energy [6–8]. Therefore, recently how to deal with the sludge reasonably and utilize the valuable compositions of sludge at the same time draws people's attention increasingly.

Although nowadays landfilling and agricultural applications are two main methods for sewage sludge disposal in many countries [9], they are being progressively abandoned due to the lack of land resources and high content of toxic substances in sludge. Comparatively, thermal treatments such as combustion, pyrolysis and gasification are more preferred due to the large reduction of sludge volume and the energy recovery [10]. However, during sludge combustion, the volatile heavy metals cannot be easily captured by gas-cleaning installation and meanwhile some polluting gases including SO_2 and NO_x are emitted, which will cause secondary pollution. Thus, in the actual processing procedure, the trace elements release and gaseous emissions are two urgent problems to be solved [11]. With regard to that, the present study was motivated.

The partitioning behavior of trace elements is influenced by many initial conditions of sludge, such as trace elements content and concentration of sulfur, chlorine, moisture, and so on. During combustion, the trace elements are vaporized from the fuel, and a series of reactions occur with the gaseous materials and ash particles [12,13]. Gerstle and Albrinck [14] have found that the concentrations of As, Ca, Hg, Zn and Pb in flue gases are increasing as combustion temperature goes up. Subsequently, the volatile metals are easy to form submicron particles during condensation, which are difficult to be captured by flue gas cleaning equipment. In order to control the escape of gaseous heavy metals from the incinerator, different reaction conditions have been studied to maximize the enrichment of heavy metals in larger particle size and thus reduce the environmental stress [15-18]. Adding solid additives has been widely recognized as a promising technology for heavy metals capture and gaseous emissions reduction, as the toxic heavy metals would react with activated solid additives by adsorption and chemical reactions [19-21]. Lopes et al. [22] have found that Pb and Cd emitted during co-combustion of sewage sludge and coal are more easily captured by dust collector. Ho et al. [23] have pointed out that in the fluidized bed the adsorption efficiency of trace elements can reach 95% owing to sorbents, and as for the adsorption efficiency of Cd and Pb, limestone is better than sand and alumina. What's more, Linak

| able 1 | | | | |
|-----------|----------|----------|--------|----------|
| roperties | of dried | d sewage | sludge | adopted. |

and Wendt [24] have found in a small semi-industrial-scale combustor Pb can be scavenged almost completely by kaolin addition. Although many efforts have been made to fix these trace elements, the mechanisms of the additives involving adsorption and chemical reaction are still under debate. Herein we employed two opposite additives, acidic kaolin and alkaline CaO, and systemically identified their effects on the partitioning behavior of heavy metals, representing two general strategies.

On the other hand, NO_x and SO_2 may cause photochemical smog and acid rain, so these gaseous emissions cannot be neglected during the sludge combustion. Normann et al. [25] and Houshfar et al. [26] have found that oxy-fuel combustion could reduce the NO_x emission. Shao et al. [27] has discovered that the average concentration of SO_2 emissions is higher in the N_2/O_2 atmosphere than that in the CO_2/O_2 . There have already many researches been conducted on co-combustion to reduce gaseous emissions [28–30]. Nevertheless, the mechanism of additives affecting the gas emission during sludge combustion is limited explicated. In the present study, the release of polluting gases would also be characterized and meanwhile the fixation effect of these two additives would be studied including the possible mechanisms.

In summary, to identify how the acidic and alkaline additives influences the distribution behaviors of hazardous elements including trace elements and polluting gases during sludge combustion, the present study was motivated. Here we provide an insight to the mechanism of two types of additives affecting the polluting materials emissions and distributions during sludge combustion. This paper mainly studies the mechanism analyses of sludge combustion processes, including kinetics and pollutant release mechanisms, which will be of great significance for people's cognation to sludge combustion. Furthermore, starting from the present study, we would provide two strategies for pollution control and possible stabilization mechanisms, which have key implications for applications.

2. Materials and methods

2.1. Raw materials

A sludge sample was collected from a municipal wastewater treatment plant in Shenzhen, China. The sewage sludge (denoted as S0) was dried in air at 105 °C for 12 h, crushed using a grinder and then sieved to a grain size below 150 μ m before experiments. Table 1 depicts the properties of dried sewage sludge, including proximate and ultimate analyses, chemical composition of sludge and concentrations of the trace elements. HHV was measured using the 6400 automatic isoperbol calorimeter (Parr Instrument Company, USA) and the oxygen content was calculated by mass difference.

In order to mitigate the environmental pollutions of sludge combustion, herein two types of additives, calcium oxide with acidity and kaolin with alkalinity, were employed. The sludge was adequately mixed with the additives using a ball mill with a mass blending ratio of 5:1 (sludge to additive). Furthermore, the samples mixed with calcium

| Proximate analyses (air dry%) | | | Ultimate analyses (air dry%) | | | | | HHV ^b (MJ/kg) | |
|--------------------------------|--------|-------------------|------------------------------|-------------|------------------------|----------------|------------------------|--------------------------|------|
| Moisture | Ash | Volatile | Fixed carbon | C | H | O ^a | N | S | 5.29 |
| 2.22 | 62.64 | 28.60 | 5.69 | 14.01 | 3.27 | 16.93 | 2.09 | 1.05 | |
| Oxide | % mass | Oxide | % mass | Trace metal | mg/kg | Trace metal | mg/kg | | |
| SiO ₂ | 32.14 | SO ₃ | 3.48 | Cu | 3.28 × 10 ³ | Cr | 1.12 × 10 ³ | | |
| Al ₂ O ₃ | 30.49 | K ₂ O | 2.03 | Ni | 571 | As | 138 | | |
| Fe ₂ O ₃ | 13.01 | MgO | 0.67 | Pb | 44.2 | Ba | 457 | | |
| CaO | 2.96 | Na ₂ O | 0.54 | Cd | 1.04 | Zn | 927 | | |

^a Calculated, O = 100 - (C% + H% + N% + S% + Ash%);

^b HHV, determined by bomb calorimeter.

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