



Research Paper

Pyrolysis and subsequent direct combustion of pyrolytic gases for sewage sludge treatment in China



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HIGHLIGHTS

- An equipment based on pyrolysis of SS and immediate combustion of pyrolytic gases as energy supplier was developed.
- The practical treatment of dry SS was carried out.
- Flue gas compositions and leaching behavior of heavy metals were investigated.
- Energy and mass balance evaluation was performed based on the treatment of 300 kg/h dry SS.
- High temperature flue gas could be utilized for drying wet SS or steam production.

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ABSTRACT

An equipment based on pyrolysis and subsequent direct combustion of the pyrolytic gases as energy supplier was developed to treat dry sewage sludge in China. In this system, the pyrolytic gases generated in the pyrolysis room were transported upward to the burning room where they were ignited to release heat energy which irradiated back to sustain pyrolysis of the sewage sludge in the pyrolysis room. Parameters such as the feeding speed and transporting speed of sewage sludge, as well as the temperatures of pyrolysis room and burning room were investigated and optimized. The compositions of the flue gas and leaching behavior of heavy metals in biochar were analyzed to assess their impact on environment. Furthermore, the energy and mass balance was investigated to figure out the efficiency of this equipment and the amount of heat energy preserved in the high temperature flue gas which could be utilized for either drying wet sewage sludge or steam production.

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1. Introduction

Sewage sludge (SS) is a semi-solid waste produced from wastewater treatment plants (WWTPs) after a series of physical, chemical and biological wastewater treatment. In 2013, the SS production in China was about 6.25 million tons (in dry solid weight), which was nearly doubled compared to the year of 2007 [1]. SS is rich in organic matter and contains many toxic substances such as heavy metals, pathogens or other microbiological pollutants, persistent organic pollutants (POPs) [1–5]. Inappropriate disposal of SS could lead to adverse environmental impacts such as public health risks and the possibility of contaminating atmosphere, soil and water resources [2].

In China, little attention has been paid to SS treatment and disposal for WWTPs. The investment ratio of SS treatment and disposal to sewage treatment is about 50% in developed countries,

however, only 8% in China from 2011 to 2015, resulting in SS accumulation in huge quantities [1]. As of 2013, more than 80% of SS was dumped improperly, the other disposal methods were sanitary landfill, land application, producing building materials, incineration, accounting for 13.4%, 2.4%, 0.24% and 0.36%, respectively [1]. The newly issued “Action Plan for Prevention and Treatment of Water Pollution” by Chinese State Council strongly emphasized that harmless SS treatment rate must reach higher than 90% in cities at prefecture level and above by the end of 2020 [6]. This action plan is bringing about great challenge and opportunity for the SS treatment market in China. Thus, environmentally and economically effective methods to treat SS are urgently needed by industry.

Anaerobic digestion and composting are biological methods for SS treatment, whereby biogas or fertilizers can be produced [7,8]. However, these biological methods can't degrade POPs significantly and show weak immobilization of the heavy metals. Consequently, agricultural use of the solid residues after biological treatment usually causes concerns about food safety and sec-

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ondary environmental pollution. Landfill of SS not only requires large area of land but also produces leachates which contain hazardous materials such as refractory organics and heavy metals, and greenhouse gas [9]. Incineration of SS can significantly reduce waste volume and recover heat energy, thereby it was supported by some people [10]. However, the releases of dioxin, NO_x , SO_2 , and heavy metals during sewage sludge incineration would cause serious air pollutions [11,12], thereby installation of relatively complicated system for the flue gas treatment is indispensable.

A complete solution of the SS problem together with recovery of energy and nutrients at acceptable costs [2] should be explored and soon tested in pilot scale. Pyrolysis has advantages such as decomposition of organic pollutants, sterilization of pathogens, stabilization of heavy metals, and large volume reduction [13–15]. The products derived from SS pyrolysis are syngas, tar, and char [12,15–18]. Syngas and tar which show a high heat value are comparable to some fossil fuel [13,16,19]. And char is considered as a promising fertilizer or soil amendment mainly because of concentrated preservation of N, P, K in char [20], water retention and soil structure improvement [21], and low bioavailability of heavy metals in char [20,21]. Furthermore, unlike combustion process, pyrolysis in an oxygen-limited atmosphere restrains the generation of air pollution materials such as NO_x , SO_2 and dioxins [11,12]. However, an external heat source is necessary if syngas and tar are collected as fuels, and additional devices are required for syngas processing and tar utilization, which would increase the treatment cost.

In this work, the developed equipment here followed the procedures that it firstly thermally decomposed the organic compounds into combustible gases, then the gases were ignited to generate heat energy and directly support the pyrolytic process, thereby syngas and tar collecting systems were unnecessary. The practical treatment process for SS was performed at various patterns of feeding speed and transporting speed. As the parameters was optimized, the operation was carried out for a long and continuous period to check the stability of the equipment. Finally, to verify the feasibility of the equipment for SS treatment, environmental evaluation and energy and mass balance evaluation were conducted.

2. Experiments and methods

2.1. Material and its characterization

The SS used in this work was collected from a WWTP in Beijing after dewatering and drying treatment. Proximate analysis was conducted to determine moisture, volatile matters, fixed carbon, and ash yields of the dry SS according to GB/T212-2008. Ultimate analysis based on GB/T30733-2014 was also carried out to obtain weight percentage of elements such as carbon, hydrogen, nitrogen, sulfur and oxygen (by subtraction). In addition, heating value of the SS was attained in accordance with GB/T213-2008.

Thermogravimetric (TG) measurements of the SS were performed on a TGA-DSC analyzer (TGA-DSC 1 STAR[®], METTLER TOLEDO). A sample weight of $10.0 \text{ mg} \pm 0.5 \text{ mg}$ was used for the TG analysis, which was conducted under a nitrogen flow rate of 15 mL/min and a heating rate of $10 \text{ }^\circ\text{C/min}$ from $25 \text{ }^\circ\text{C}$ to $900 \text{ }^\circ\text{C}$. The thermogravimetric and differential thermogravimetric (TG-DTG) data were used to characterize the pyrolysis behavior of the SS, as well as to provide estimates of their kinetic parameters.

CSD IDEA (Beijing) Environmental Test & Analysis Co., Ltd. was entrusted with the measurement of chemical properties of flue gas. While the POPs in the flue gas, especially dioxins (PCDD/Fs) was measured by Center for Environmental Quality Test, Tsinghua University, according to Chinese National Standard HJ77.2-2008

(Ambient Air and Flue Gas Determination of Polychlorinated Dibenzop-dioxins (PCDDs) and Polychlorinated Dibenzofurans (PCDFs) Using Isotope Dilution HRGC-HRMS) [22].

In addition, heavy metals in SS and its biochar were analyzed with standard methods: flame atomic adsorption spectrometry (air acetylene flame) for Cu, Cr and Ni, graphite furnace atomic adsorption spectrometry for Pb and Cd, atomic fluorescence spectrophotometry for As and Hg. With respect to the leaching behavior of heavy metals in SS and its biochar, it was studied according to the modified Chinese National Standard HJ/T299-2007 (Solid Waste-Extraction Procedure for Leaching Toxicity-Sulphuric Acid & Nitric Acid Method) [23]. Pretreatment of samples by microwave-assisted acid digestion was performed, then Cu, Pb, Zn, Cd, Cr and Ni were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS). Arsenic was determined by atomic fluorescence spectrophotometry.

2.2. Equipment and procedures

The equipment shown in Fig. 1 is mainly composed of six sections, namely feeding section, pyrolysis (carbonation) section, burning section, re-burning section, purification section of the flue gas and discharging section of bio-char. It should be noted that the pyrolysis section must to be connected with the burning section, and their horizontal length is 3.9 m. The cross sections of the pyrolysis room and the burning room are an inverted trapezoid ($1.8 \text{ m} \times 1.2 \text{ m} \times 1.8 \text{ m}$) and a rectangle ($1.8 \text{ m} \times 1.8 \text{ m}$), respectively. The installed air-blowers for the pyrolysis room, burning room and re-burning room are located as shown in Fig. 1. The thermocouples of pyrolysis room and burning room are located as shown in Fig. 2.

The SS can be automatically transported by a conveyor into the hopper, which connects to the pyrolysis room. The main shaft inside the pyrolysis room is fixed with paddles and protected from over-heating with tap water cooling system, its rotation stirs and transports the SS toward the discharging area. Both condensable and non-condensable gases released during pyrolytic process rise upward and are ignited in the burning room in the presence of air. The combustion generates a huge amount of heat, which irradiates back to support the SS pyrolysis. Considering that the pyrolytic gases are burned incompletely in the burning room, re-burning room has been designed for further burning of combustible gases. Thereafter, the exhaust gas flows into the cyclone in which dust and gas can be properly separated, and then be emitted. The solid residue after pyrolysis is bio-char. It is transported and pushed to the screwing output conveyor by the paddles of the main shaft. To cool down the bio-char, tap water is also circulating within the screwing shaft of output conveyor.

The rotating speed of the screwing shaft in the hopper and the main shaft in the pyrolysis room are controlled by a Control Panel. Their rotating speed is proportional to their Hz numbers. By increasing the Hz number, the shafts will rotate faster, which means more SS is feeding into the pyrolysis room and the residence time for SS pyrolysis is shortened. The quality of pyrolytic products are closely related to the treated amount of SS per hour and its residence time in pyrolysis room. Therefore, the Hz number of the two shafts should match each other to attain pyrolytic products of good quality. Three air-blowers have been settled respectively for the pyrolysis room, burning room and re-burning room. The quantity of air input is controlled by series of valves. During equipment startup, auxiliary fuel such as diesel oil is indispensable to heat up the equipment. However, as the temperature reaches to the requirement, burning of diesel oil will be automatically turned off since combustion of pyrolytic gases can sustain the SS pyrolysis process, and the quantity of air input into the pyrolysis room should be adjusted so that pyrolysis of SS occurs under oxygen-absent or oxygen-limited

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