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Combustion behavior, emission characteristics of SO_2, SO_3 and NO, and in situ control of SO_2 and NO during the co-combustion of anthracite and dried sawdust sludge

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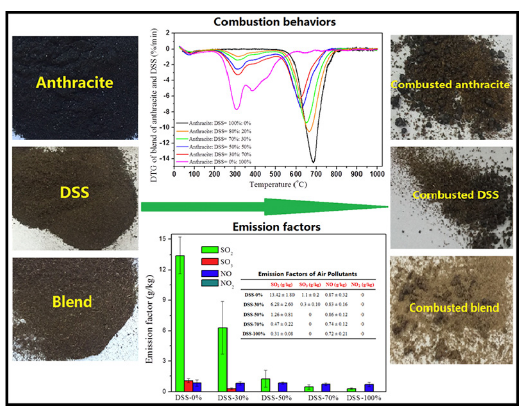
HIGHLIGHTS

- The co-combustion behavior of anthracite and dried sawdust sludge (DSS) was studied.
- DSS improved the ignition of anthracite and accelerated the burning rate.
- DSS decreased the SO_2 emission factor but had no function in mitigating NO emission.
- The removal efficiencies of SO_2 and NO reached 100% and 95% by two control methods.
- The removal products were determined as sulfate, sulfite and nitrate by IC.

GRAPHICAL ABSTRACT



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ABSTRACT

The combustion behaviors of anthracite and dried sawmill sludge (DSS) were studied using thermogravimetric analysis (TGA) and derivative thermogravimetric analysis (DTG). DSS was found to be a promoter for anthracite combustion, the addition of DSS in anthracite decreased the burnout temperature and time. But DSS caused the rapid releases of SO_2 and NO in the initial combustion stage. In overall, the increasing of DSS significantly decreased the emission factor of SO_2 from 13.42 ± 1.80 to 0.31 ± 0.08 g/kg; while the emission factor of NO was not obviously changed and stable at 0.7–0.8 g/kg in all cases. The oxygen-rich atmosphere was helpful for the rapid and sufficient combustion of blend; the oxygen-lean atmosphere delayed the combustion process and slowed down the releases of SO_2 and NO. The increasing combustion temperature improved the anthracite combustion, and the emission factors of SO_2 and NO were all increased with the temperature increasing. 900 °C was found to be the best combustion temperature for NO generation. SO_3 was detected in the combustion of anthracite under 21% and 30% of O_2. Two promising ways for control of SO_2 and NO were provided: 1) urea-fuel mixture combustion combined with the post-

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combustion wet absorption by Na_2CO_3; 2) post-combustion wet absorption by $NaClO/Na_2CO_3$. The removal efficiencies of SO_2 and NO could reach 100% and over 95% respectively. The removal products were determined as sulfate, sulfite and nitrate by IC, with no toxic byproducts being produced.

1. Introduction

Energy utilization of sludge has been received extensively attention as a consequence of this process can not only solve the environmental problems caused by sludge but also enable the efficient reuse of energy from sludge (Nadziakiewicz and Kozioł, 2003; Dong et al., 2015; Kijo-Kleczkowska et al., 2016; Sahu, 2014; Zhang et al., 2015). The sludge contains large amounts of salts, heavy metals and organic pollutants those will significantly harm the humans and environment if not treated properly. The conventional methods for the energy utilization of sludge mainly include co-combustion with coal (Nadziakiewicz and Kozioł, 2003; Dong et al., 2015; Kijo-Kleczkowska et al., 2016; Sahu, 2014), anaerobic digestion with producing methane (Zhang et al., 2015), low-temperature pyrolysis to generate fuel oil (Reckamp et al., 2014), gasification to form combustible gas (Gil-Lalaguna et al., 2014), and slurry synthesis of coal sludge (Wang et al., 2012). Among these methods, co-combustion of sludge and coal is desirable for treating sludge as a result of the superiorities of decreasing the sludge volume, eliminating the pathogenic organisms, thermally destroying the organic/toxic components, disposing the waste materials with air pollution control devices (APCDs), and eliminating the need for waste incinerators.

A lot of works have been conducted to investigate the behaviors of co-combustion of coal and sludge. Dong et al. (2015) carried out the co-combustion of tannery sludge and bituminous coal in a 220 t/h fluidized bed boiler, and found that the tannery sludge showed a higher reactivity than bituminous coal, the co-combustion was favorable for coal ignition. Coimbra et al. (2015) investigated the feasibility of co-combustion of pulp mill sludge and bituminous coal, it was demonstrated as a satisfactory method for treating sludge, meanwhile reducing the emissions of CO_2 and NOx. Yang et al. (2016a) conducted the co-combustion of dried sewage sludge pellets and coal gangue. Liao and Ma (2010) and Hu et al. (2015) conducted the co-combustion of semi-anthracite and paper mill sludge, and revealed the synergistic effects between the two materials during the co-combustion process. Chen and Wu (2009) verified the char yield was linear relation to the biomass proportion during the co-combustion of rice husks and coal. Gil et al. (2010) studied the co-combustion of pine sawdust and coal, but no obvious interactions between biomass and coal were observed. The above studies demonstrated the co-combustion of sludge and coal was a secure outlet so long as the sludge proportions and operational conditions could be controlled appropriately. But prior to the real application of co-combustion technology, we should study the emission characteristics of air pollutants, in addition to the combustion performance, this is because a series of terrible air pollution incidents have been happened in China, and Chinese government has issued the ultra-low emission standards of SO_2 and NOx for industrial manufacture process (SO_2 < 35 mg/m^3, NO < 50 mg/m^3) (Hao et al., 2018a). Hence, the research on the emission characteristics of air pollutants as well as the in situ control of air pollutants during the co-combustion process is of great significance. Recently, some papers had studied the emission characteristics of SO_2 (Yang et al., 2016a; Zhang et al., 2016; Li et al., 2010; Wu et al., 2011), NO_x (Dong et al., 2015; Yang et al., 2016a; Zhang et al., 2016; Li et al., 2010; Wu et al., 2011), HF (Zhang et al., 2016), HCl (Zhang et al., 2016; Wu et al., 2011; Hilber et al., 2007), dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) (Zhang et al., 2016; Liu et al., 2006), particles and heavy metals (Yang et al., 2016a) during the co-combustion of municipal sludge, tannery sludge (Dong et al., 2015), sewage sludge (Yang et al., 2016a), pickling sludge (Zhang et al., 2016), solid recovered fuel (Wu et al., 2011), hazardous wastes (Liu et al., 2006) and coal. But their common problems were the low calorific values that may affect the stable combustion of coal and the relative high contents of hazards.

Dried sawmill sludge (DSS) has abundant source in China, Canada, Japan and Russia. Compared with the above sludges, DSS, as a biomass-based and relative green sludge, seems more suitable to be adopted as a combustion improver for coal since it has a relatively high calorific value but a low ignition temperature. While DSS is intrinsically different from the raw wood, it is actually the removal product from wastewater treatment process, containing a large amount of adsorbed water, salts and particulate matters, and its pyrolysis behavior and the emission characteristics of air pollutants are different from those of biomass-fuel. Hence, in order to utilize DSS as an alternative fuel source, we should firstly investigate the combustion behavior of DSS under different proportions with coal.

Anthracite is a primary energy source with a high calorific value but a very high ignition temperature. Hence, the combination of DSS and anthracite is complementary. As aforementioned, the emission characteristics and emission factors of air pollutants are crucial for the application of co-combustion technology, so another research point in this paper is to investigate these relations and provide the key data for help designing and arranging APCDs. Not only to this, the in situ control of SO_2 and NO during and after combustion was also carried out, and some basic data and useful suggestions were provided. Finally, the removal products were also analyzed by Ion Chromatography (IC), in order to support the speculation on the reaction mechanism.

2. Experimental section

2.1. Materials and analysis

The anthracite and DSS used in the experiments were obtained from Longyan coal district and some sawmill in Fuzhou, Fujian province. Anthracite and DSS were firstly dried at 105 °C for at least 24 h, then both of anthracite and DSS were ground into powders with the particle size of 200-mesh.

The proximate analyses were determined by electro-thermostatic blast oven (DGG-9123AD, China), muffle furnace (5E-MF6000, China), and proximate analysis instrument (5E-MAG6700, China). The ultimate analyses were determined using an element analyzer (5E-CHN2000, China). And the analyses data of anthracite and DSS are shown in Table 1. As Table 1 shows, the C, S and lower heating value of DSS are lower than those of anthracite, but those contents of H, O, N, ash, water and volatile in DSS are higher. It is worthy to point out that the contents of C, H and O, and the lower heating value of DSS are far higher than other sludge such as municipal sludge (Zhang, 2017) and electroplating sludge (Zhang, 2017). Besides, the contents of heavy metals in DSS were also less. Hence, it is reasonable to adopt DSS as a combustion improver for anthracite.

TG and DTG analyses of anthracite, DSS and the blends with different mass proportions of DSS (0, 30, 50, 70 and 100%) were performed in a thermo-gravimetric analyzer (Mettler-Toledo TGA/SDTA851e). The air dried samples of 20 mg were used for each analysis. N_2 or air was used as a reaction gas with a flow rate of 80 mL/min. The heating rate was set to 10, 20 and 30 °C/min, and the samples were heated from 25 to 1000 °C.

2.2. Equipment and procedures

The combustion experiments of anthracite, DSS and their blends were carried out in a horizontal tube furnace (SK-2-13, Yong Guang