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Sludge Acts as a Catalyst for Coal during the Co-Combustion Process Investigated by Thermogravimetric Analysis

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Abstract: Sewage sludge in China has the characteristics of low organic content and low heating value compared with other developed countries. Self-sustaining combustion of Chinese sludge cannot be achieved when the moisture content is high. Co-combusting a small amount of sludge in the existing coal-fired boilers is a usual sludge disposal method in China. Thermogravimetric (TG) and differential scanning calorimetry (DSC) analysis of a bituminous coal, three different sewage sludges, and their blends have been carried out. Fitted curves by linear calculation and actual curves of blends were compared to study the interaction between sludge and coal in their co-combustion process. The results indicate that the interaction between the two fuels takes place during the devolatilization and combustion period. Sludge acts as a catalyst for coal during the co-combustion process because of the large amount of inorganic salts contained in the sludge. Co-combustion of coal and sludge is more efficient than single burning of the two fuels.

Keywords: catalyst; coal and sludge; co-combustion; thermogravimetric (TG) analysis

1. Introduction

With the acceleration of the urbanization process in China, the wastewater treatment capacity has gradually increased, which generates a great deal of sludge. It is estimated that annual sludge production in China will exceed 60 million tons in 2020 [1]. Sewage sludge is comprised of concentrated pollutants produced in the sewage treatment process, which contains heavy metals, pathogenic microorganisms, and organic pollutants. Inattentive dumping of sludge will cause serious secondary pollution [2].

Combustion of sewage sludge has been a widely used solution for its treatment and disposal. Compared with other developed countries, sludge in China has the characteristics of low volatiles content and low heating value. Self-sustaining combustion of Chinese sludge cannot be achieved when the moisture content is high. It often needs to be co-burned with coal.

Co-combustion of sludge and coal has attracted extensive attention of researchers in recent years. Zhang et al. [3] concluded that the performance of co-combustion of sewage sludge with coal is sometimes better than the single burning of coal investigated by thermogravimetric (TG) analysis and drop tube furnace experiments. Hu et al. [4] used the computational fluid dynamics method to simulate the co-combustion of coal with printing and dyeing sludge in a pulverized coal power plant. The optimal sludge content, moisture content and secondary air distribution scheme were determined by the analysis of co-combustion characteristics and NO_x emissions. Lopes et al. [5] have performed some studies on ash partitioning, the formation of gaseous pollutants, and heavy metals behavior in the co-combustion process of sludge and coal. Karlsson et al. [6] discovered that co-combustion of sludge in a fluidized bed boiler can reduce high-temperature corrosion. Zhang et al. [7] showed that

co-combustion of CaO-condition sludge and coal is a suitable and promising technology for clean disposal of sludge. Zhu et al. [8] conducted their experiments on a pilot-scale circulating fluidized bed. The results showed that co-combustion of sludge and coal possessed a series of advantages, such as stable combustion, high efficiency, and low emission of pollutants. Tan et al. [9] co-fired sludge in an existing coal-fired utility boiler, which showed co-firing 10% sludge with a moisture content ranging from 40% to 56% is optimal, and the annual profit is remarkable. Chen et al. [10] investigated the emission characteristics of polycyclic aromatic hydrocarbons and their toxic equivalent concentration during coal/sludge co-combustion process in a laboratory-scale drop tube furnace at temperatures in the range of 950–1250 °C. The results demonstrated the pollutants generated from 30/70 (sludge/coal) co-combustion were lower than their linear calculations. These references show a series of advantages and the feasibility of co-burning coal with sewage sludge.

Some scholars have also started to explore the feasibility of co-burning sludge with other biomass. Rong et al. [11] studied the combustion characteristics and slagging during co-combustion of sludge and rice husks. The ratio of sludge in the blends cannot exceed 30% according to the results. Liu et al. [12] quantified the mass loss response of co-burning sludge and water hyacinth. Lin et al. [13] used TG analysis to investigate the co-combustion behaviors between sludge and oil shale. The blend with 10% sludge mixing ratio showed the best fuel performance. Xie and Ma [14] also used TG analysis to study the thermal behavior of the co-combustion between sludge and rice straw. Additionally, other waste and biomass such as animal waste [15], palm kernel shell [16], and human feces [17] are also considered to be utilized by co-combustion technology.

TG analysis is a commonly used method to see the combustion characteristics of fuels. Several similar works about co-combustion of sludge and coal have been published. Among these studies, some are focused on the kinetic analysis and combustion characteristic parameters. Liao et al. [18] and Otero et al. [19] indicated the combustion performance of the blends with low percentages of sludge (<10%) is similar to that of coal. Magdziarz and Wilk [20] discovered coal can be burned beneficially with biomass and sludge. Some used other equipment linked to TG like mass spectrometry, to investigate the gas emissions. Otero et al. [21] discovered that the CO₂ emission pattern is very similar to the corresponding differential thermogravimetric (DTG) curve. In their later study, they found that when the sludge percentage in the blend is 10% or less, emission of CO₂ is very similar to that corresponding to the coal, while the emission of NO₂ is slightly higher, and that of SO₂ is lower [22]. There are a few discussions about the interaction between coal and sludge during their co-combustion process in recent studies. Ninomiya et al. [23] carried out experiments in a laboratory-scaled drop tube furnace to understand the interaction between sludge and coal. The results showed the obvious interaction that the devolatilization of mixing fuel occurred quickly and the combustion of both char and evolved volatiles progressed almost completely. However, some scholars think there is no interaction between the two fuels [22,24].

As co-combustion of coal and sludge is a feasible method to realize clean disposal of sewage sludge, the interaction between these two fuels during their co-combustion process needs to be thoroughly studied. TG and differential scanning calorimetry (DSC) analysis of a bituminous coal, three different sewage sludges, and their blends have been carried out in this work. A non-traditional analysis method for TG curves has been put forward. Fitted curves by linear calculations and actual curves of blends were compared to study the interaction between two, or several materials, in a certain temperature-programmed process. This research provides essential and fundamental data for the feasibility and optimization of co-burning sewage sludge in existing coal-fired power plants.

2. Experimental

2.1. Materials

A bituminous coal (labeled as C) and three different sewage sludges (labeled as S₁, S₂, S₃) have been employed in this research. The bituminous coal was collected from the coal storage of the Jiaxing

electric power plant, Zhejiang Province. S_1 comes from the Jiaying sewage treatment plant, and it is generated from a mixing treatment of both municipal sewage and industrial wastewater. S_2 is tannery sludge from a tannery factory, and S_3 is municipal sludge from the Pinghu wastewater treatment plant in Shenzhen, Guangdong Province. All the samples were dried in an electric drying oven (fan blown type) at 105 °C for 12 h, after which they were milled and meshed to small particles with a size less than 154 μm .

The proximate and ultimate analysis results and higher heating values of these samples are shown in Table 1. The volatiles content in average of these three Chinese sludges is only 40.08%, much lower than that of European countries (60% on average) [25].

Table 1. Proximate, ultimate analysis results, and higher heating values of the samples.

Samples	Sludges			Coal
	S_1	S_2	S_3	
Proximate analysis (wt %)				
Moisture	5.16	3.70	3.37	3.55
Volatiles	40.74	38.78	35.76	30.69
Ashes	52.76	56.93	56.87	13.20
Fixed carbon	1.34	0.59	4.00	52.56
Ultimate analysis (wt %)				
C	20.98	15.31	16.82	67.97
H	3.31	2.49	3.36	4.04
N	2.50	1.50	3.23	1.05
S_{total}	4.32	0.81	1.03	0.48
O	10.97	19.26	15.32	9.71
Higher heating values (MJ/kg)				
HHV	9.251	5.934	8.446	21.935

2.2. TG Analysis

TG analysis was performed in a NETZSCH analyzer, model STA 449 F3 (NETZSCH, Selb, Germany). It is able to simultaneously collect TG-DSC signals. The experiments were carried out at atmospheric pressure and temperatures ranging from room temperature to 1000 °C under an air atmosphere. The flow rate of the air was fixed at 80 mL/min and a ca. 10 mg sample was used for each experiment. Each sample was put in an Al_2O_3 crucible when tested. Tests were carried out mainly for S_1 , C, and their sludge-coal blends, while the analysis of S_2 and S_3 sludge-coal blends are a comparison and verification. Test conditions regarding dried sludge weight percentages of blends and corresponding heating rates are listed in Table 2.

Table 2. Test conditions list.

Samples	Dried Sludge Weight Percentages (%)	Heating Rate (°C/min)
S_1 and C	0, 10, 30, 100	20
	0, 5, 10, 20, 30, 100	30
	0, 10, 30, 100	40
S_2 and C	0, 10, 30, 100	30
S_3 and C	0, 10, 30, 100	30

2.3. Data Analysis Methods

Assuming there is no interaction between sludge and coal in the co-combustion process, the combustion of sludge and coal in the mixed sample can be regarded as two separate reactions.

On this assumption, the TG, DTG and DSC analysis curves of a sludge-coal blend can be represented according to their respective curves and sludge weight percentage of the mixed sample. The curves obtained by this method are called fitted curves and described by Equations (1)–(3),

$$TG_{f,M_S} = TG_S \cdot M_S + TG_C \cdot (1 - M_S) \quad (1)$$

$$DTG_{f,M_S} = DTG_S \cdot M_S + DTG_C \cdot (1 - M_S) \quad (2)$$

$$DSC_{f,M_S} = DSC_S \cdot M_S + DSC_C \cdot (1 - M_S) \quad (3)$$

where M_S is the sludge weight percentage of the mixed sample, subscript f represents the fitted curve and subscript S, C represents the sludge and coal, respectively. In fact, the interaction between sludge and coal exists in the co-combustion process. Therefore, the differences between the fitted curves and the practical analysis curves show the effects of the interaction.

3. Results and Discussion

3.1. Combustion Characteristics of Coal and Sludge Samples

Coal and sludge have totally different combustion characteristics. On the other hand, in spite of having some differences, sludge samples from different sources perform similarly. TG and DTG curves for coal and sludge samples are shown in Figure 1. The whole temperature-programmed processes of samples can all be divided into three periods. Due to the moisture content, the four DTG curves all have a small weight loss peak in their first periods.

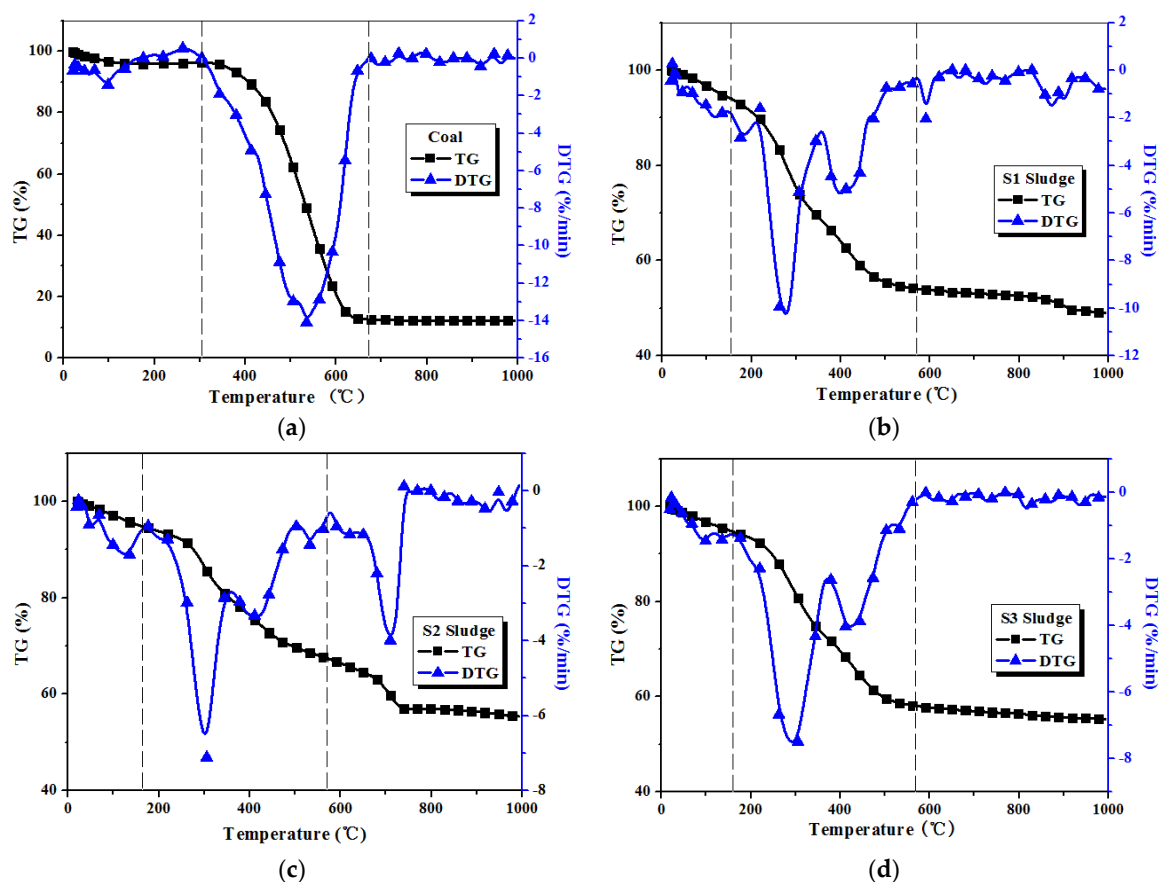


Figure 1. Thermogravimetric (TG) and differential thermogravimetric (DTG) curves for coal and sludge samples (heating rate = 30 °C/min) ((a) coal; (b) S₁; (c) S₂; (d) S₃).

The second period, called the devolatilization and combustion period, of three sludge samples begins at about 180 °C and ends at about 550 °C. There are two peaks in this period of all the sludge samples, which can be regarded as the devolatilization of two kinds of main organic components in the sludge. In a previous work on paper sludge, the first peak was recognized as the decomposition of hemicellulose and cellulose, while the second corresponded to the decomposition of lignin [26]. The first peak is at about 300 °C and the second one is at about 440 °C. Due to the higher volatile content, the corresponding weight loss rate of S_1 (10.9% min⁻¹ and 5.1% min⁻¹) is a slightly larger than S_2 (7.2% min⁻¹ and 3.6% min⁻¹) and S_3 (7.7% min⁻¹ and 4.0% min⁻¹). However, the combustion characteristic of coal is very different from those of the sludge samples. The second period of coal begins at 300 °C and ends at 680 °C. Both the temperatures are much higher than that of the sludge samples, which indicates the combustion of coal requires a higher temperature than sludge. There is only one weight-loss peak in this period of coal, with a maximum weight loss rate of 13.9% min⁻¹ at 540 °C. This indicates that the combustion temperature of sludge is much lower than that of coal, but the reactions are more complex.

During the last period, called the burnout period, coal and S_3 have almost no reactions, while S_1 and S_2 perform differently because of their different sources. In the case of S_1 , a small peak occurs at about 880 °C with a weight loss rate of 1.8% min⁻¹. Some scholars put forward that the combustion of fixed carbon attributes to the small weight loss peak in the burnout period of the sludge combustion process [20]. However, coal and S_3 with higher fixed carbon yield have no peak in the last period and the decomposition temperature of pure calcium carbonate, which may exist in sludge, is 825 °C close to 880 °C. Thus, the decomposition of carbonates may cause the weight loss peak of S_1 . In the case of S_2 , a significant peak occurs at about 720 °C with a weight loss rate of 3.9% min⁻¹. Due to the high reaction intensity, the weight loss peak cannot be the decomposition of carbonates or the combustion of fixed carbon. It may be the decomposition of some organic polymer and the devolatilization that is attributed to this weight loss peak. It is also the reason why S_2 has higher volatile content than S_3 , but has a lower reaction intensity in the second period.

3.2. Comparison between Actual and Fitted DTG Curves for Coal-Sludge Blends

As mentioned above in Section 2.3, fitted curves by linear calculation and the actual curves of the blends were compared to investigate the interaction between coal and sludge during the co-combustion process. The DTG results for coal-sludge (S_1) blends are shown in Figure 2. When the sludge weight percentage of the blend is 5%, there is no obvious difference between the actual and fitted curves because the quantity of sludge is too small to influence the whole combustion process. When the sludge weight percentage of the blend is 10% or more, some differences between the actual and fitted curves has appeared, which means the interaction has occurred. Interaction between coal and sludge mainly takes place during the devolatilization and combustion period. Except the figure of the 5% blend ratio, the other figures show the coal reaction peak of the actual curve is sharper than that of the fitted curve. The maximum weight loss rates of the actual curves are all larger than that of the fitted curve. This means, in the actual process, the combustion of mixed fuel is quicker and more violent. Since the main reaction peaks of the curves are the coal combustion peak compared with Figure 1, it can be inferred that the addition of sludge can catalyze and promote the combustion of coal. Sludge acts as a catalyst for coal during the co-combustion process.

In order to see the influence of the heating rate to the interaction between coal and sludge, the tests with different heating rates have been carried out. All the figures have the same characteristics as Figure 2. The maximum weight loss rates of the fitted curves and the actual curves have been listed in Table 3. When the heating rate or sludge weight percentage increases, the differences between the actual and fitted curves become more obvious. In actual incineration furnaces, the heating rate is much higher than the test, so the interaction between the two fuels cannot be neglected.

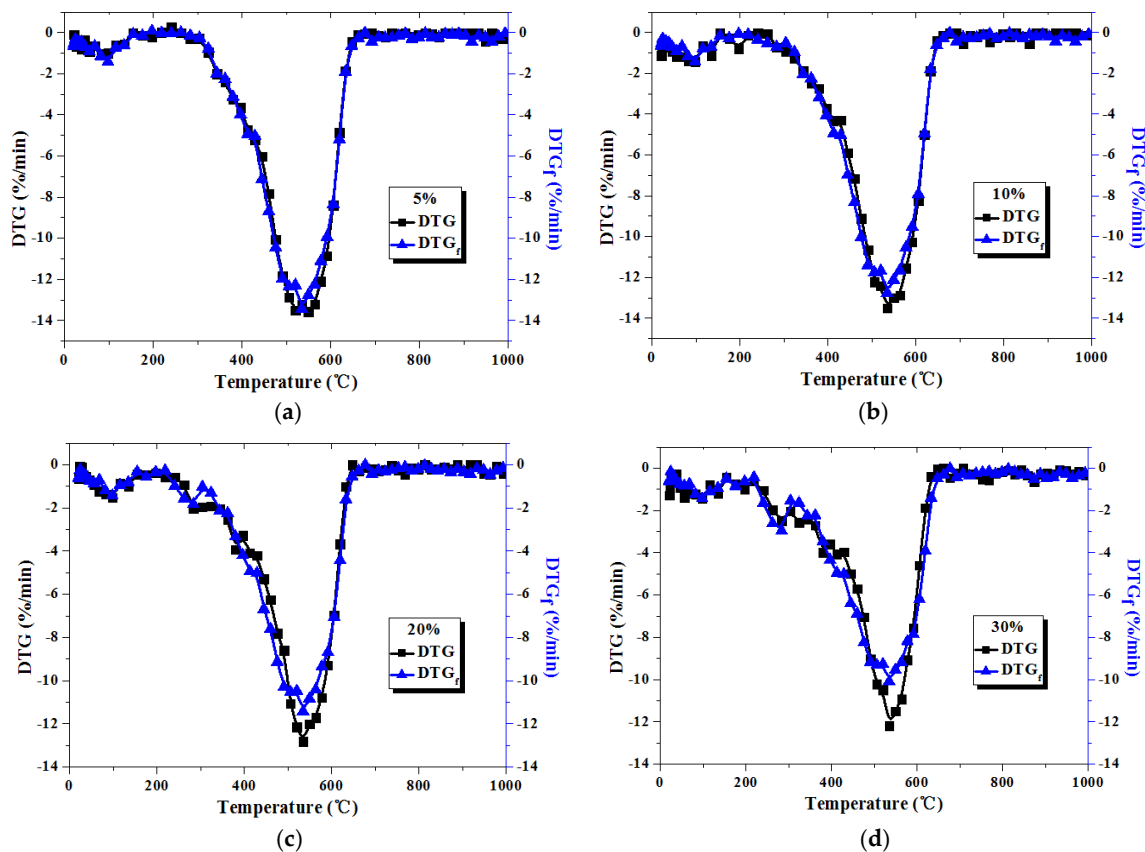


Figure 2. Actual and fitted DTG curves for coal-sludge (S_1) blends of different dried sludge weight percentages (heating rate = $30\text{ }^\circ\text{C}/\text{min}$) ((a) 5%; (b) 10%; (c) 20%; (d) 30%).

Table 3. Comparison of the maximum weight loss rate of the fitted and actual curves with different heating rate.

Dried Sludge Weight Percentage (%)	Heating Rate ($^\circ\text{C}/\text{min}$)	Maximum Weight Loss Rate of Fitted Curves (%/min)	Maximum Weight Loss Rate of Actual Curves (%/min)	Change Rate (%)
10	20	10.641	11.075	4.08
	30	12.765	13.511	5.84
	40	14.635	15.777	7.80
30	20	8.396	9.264	10.34
	30	10.284	12.088	17.54
	40	11.471	13.915	21.31

The inorganic components' content of coal and S_1 were measured by X-ray fluorescence equipment (XRF, Thermoscientific ARL ADVANT'X IntelliPowerTM 4200) in order to see the causes of the catalytic action. As shown in Figure 3, the inorganic composition of sludge is more complex than that of coal. All the components' content of sludge is much greater, except TiO_2 . Coal has low content of MgO , P_2O_5 , K_2O , and Na_2O , while each of them have certain amounts in sludge. Among the compounds, some have catalytic effects on the combustion of coal. Potassium, sodium, and calcium catalysts are the most common catalysts for catalytic gasification of coal because of its low price and good catalytic effect. Yeboah et al. [27] and Sheth et al. [28] selected Na_2CO_3 and K_2CO_3 to study their performance on coal catalytic gasification while calcium compounds were chosen by Jiang et al. [29]. In addition to alkaline and alkaline-earth metal compounds, oxides and salts of transition metals, like Fe, also have catalytic action, which was shown in the studies of Domazetis et al. [30], Popa et al. [31],

and Zhang et al. [32]. Yeboah et al. [27], Fan et al. [33], and Monterroso et al. [34] discovered that the composite catalyst combined with these three kinds of catalysts has a higher catalytic efficiency. Thus, the sludge contains a large amount of catalyst for coal gasification. Though the atmosphere of coal combustion and gasification is different, the addition of sludge accelerates the devolatilization of coal in its combustion process according to the catalytic mechanism. This is the reason why the maximum weight loss rates of the actual curves are all larger than that of the fitted curves.

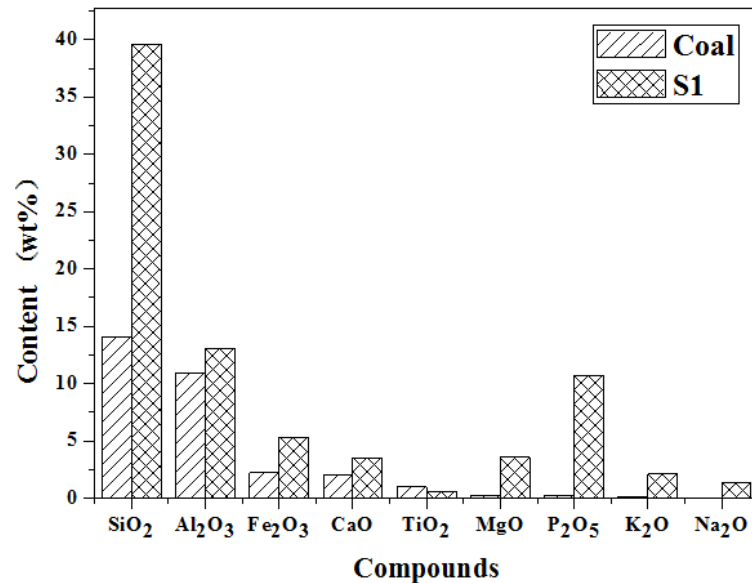


Figure 3. Inorganic compositions of coal and S₁.

In order to see if other kinds of sludge have the same effects on the coal during the co-combustion process, the tests for the same coal and sludge of different sources have been carried out, as shown in Figures 4 and 5. It can be seen that all the figures also have the same characteristics as Figure 2. The maximum weight loss rates of all actual curves are larger than that of the fitted curves. Therefore, if the problem of increased pollutant emissions, probably caused by the addition of some high-sulfur and high-nitrogen sludge, can be solved by the increasingly improved exhaust treatment facilities and clean combustion processes, co-combusting a small amount of sludge in some small existing co-fired boilers could be a good choice for sludge disposal and treatment in China. This can improve the combustion efficiency of the boiler at the same time.

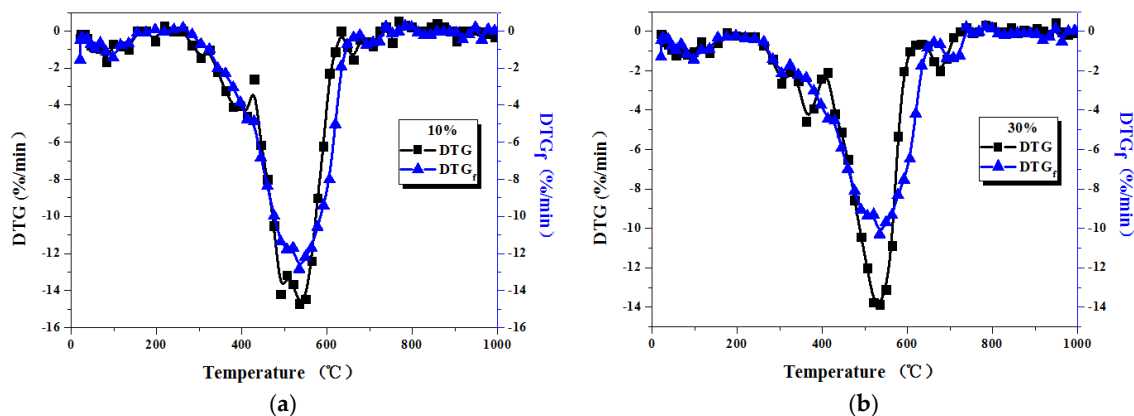


Figure 4. Actual and fitted DTG curves for coal-sludge (S₂) blends of different dried sludge weight percentages (heating rate = 30 °C/min) ((a) 10%; (b) 30%).

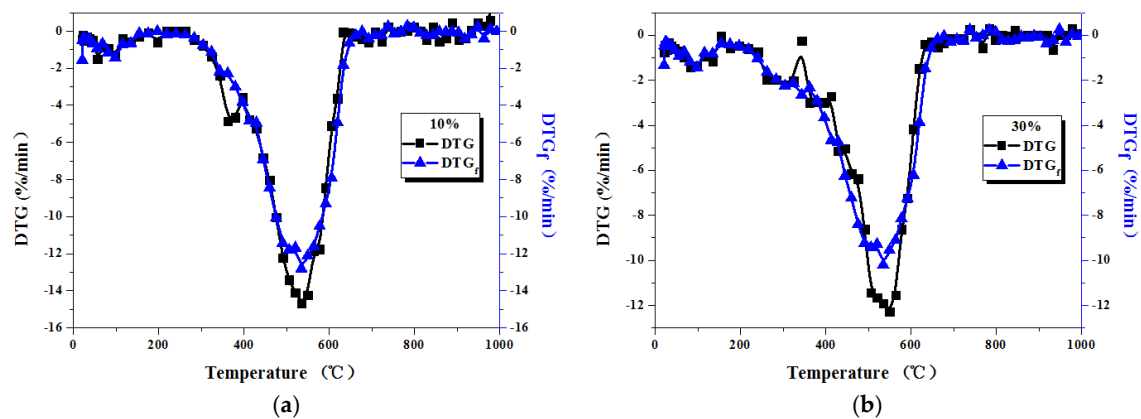


Figure 5. Actual and fitted DTG curves for coal-sludge (S_3) blends of different dried sludge weight percentages (heating rate = $30\text{ }^\circ\text{C}/\text{min}$) ((a) 10%; (b) 30%).

As mentioned in Section 3.1, a small peak occurs in the DTG curve at about $720\text{ }^\circ\text{C}$ in the case of S_2 . In Figure 4, a small weight loss peak also occurs in the high-temperature region, which is superimposed with the coal reaction peak and the high-temperature reaction peak of S_2 . Thus, the temperature of the maximum weight loss rate of this small peak is about $680\text{ }^\circ\text{C}$, slightly lower than $720\text{ }^\circ\text{C}$. The figures show that the small peak of the actual curve appears earlier. This indicates that the coal can accelerate the decomposition of the polymer material in the sludge during the co-combustion process.

3.3. Comparison between Actual and Fitted DSC Curves of Coal-Sludge Blends

The DSC results for coal and sludge (S_1) with their DTG curves are shown in Figure 6. Since the combustion process of coal is mainly the reaction of fixed carbon, the heat release of coal is synchronized with its weight loss. While the combustion process of sludge is mainly the decomposition of organic materials, and then the burning of combustible gases, the heat release of sludge is slightly later than its weight loss. As a result, the temperature of maximum energy release during coal combustion is $540\text{ }^\circ\text{C}$ the same as the temperature of the maximum weight loss rate. The temperature of the maximum energy release during S_1 sludge combustion is $300\text{ }^\circ\text{C}$ higher than the temperature of maximum weight-loss rate ($280\text{ }^\circ\text{C}$). There is also an endothermic process for a short time period at around $400\text{ }^\circ\text{C}$ which could be related to the decomposition of the second organic compound. In addition, two other kinds of sludge (S_2 , S_3) also have the same properties. However, for the sake of brevity, their curves are not shown in the paper.

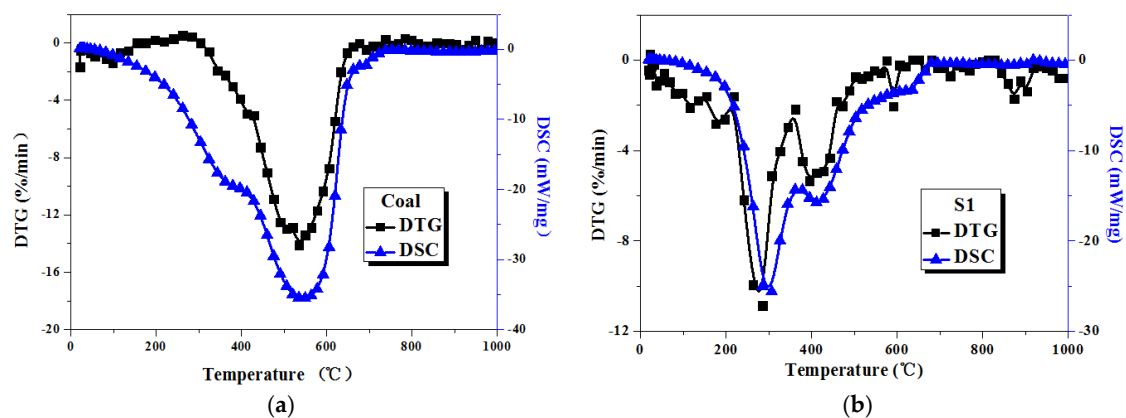


Figure 6. Differential scanning calorimetry (DSC) and DTG curves for coal and S_1 sludge sample (heating rate = $30\text{ }^\circ\text{C}/\text{min}$) ((a) coal; (b) S_1).

Similarly, the actual and fitted DSC results for coal-sludge (S_1) blends are shown in Figure 7. The differences between actual and fitted DSC curves have also proved that the addition of sludge catalyzes and promotes the combustion of coal. The maximum heat release rates of actual figures are larger than that of the fitted figures. Except the peak, there is little difference between actual and fitted DSC curves. Thus, the actual total heat release of obtained by integrating the DSC curves is slightly larger than that of the fitted value, which indicates co-combustion of coal and sludge is more efficient than single burning of the two fuels.

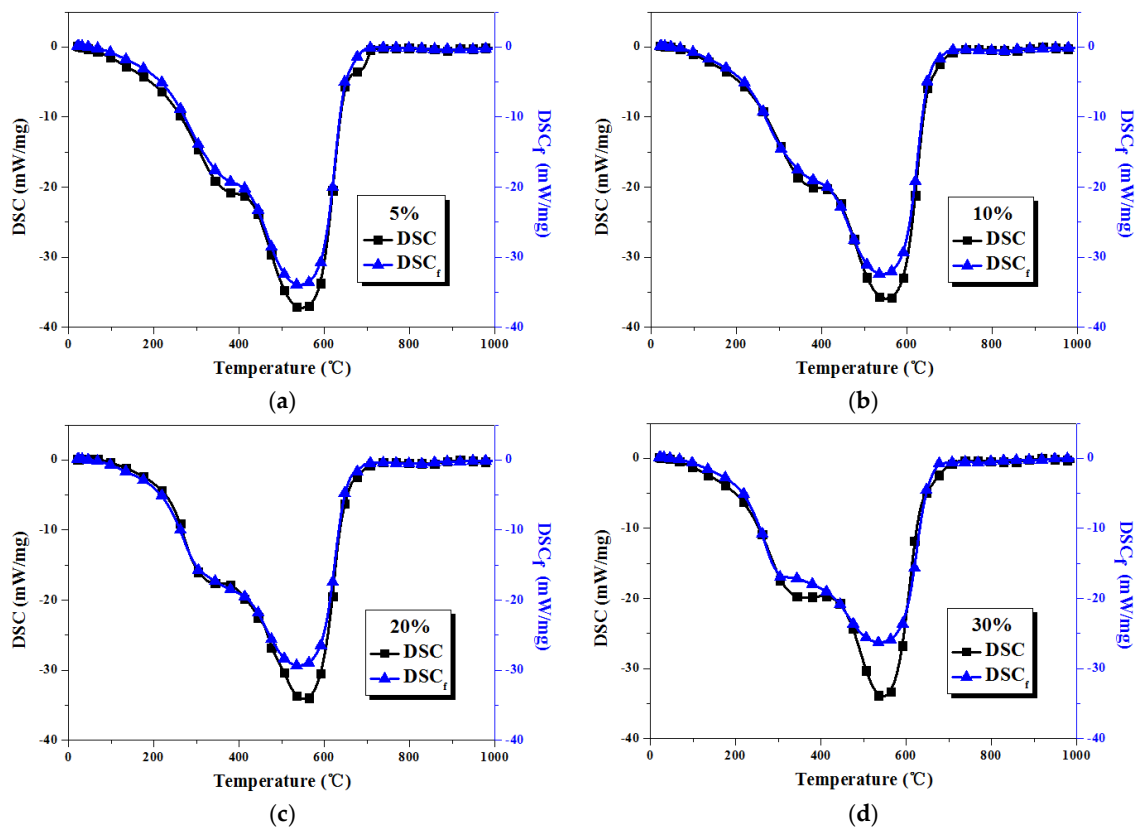


Figure 7. Actual and fitted DSC curves for coal-sludge (S_1) blends of different dried sludge weight percentages (heating rate = 30 °C/min) ((a) 5%; (b) 10%; (c) 20%; (d) 30%).

4. Conclusions

TG analysis of coal, sludge of different sources, and their blended samples have been carried out in this study. The TG and DTG curves show that coal and sludge have totally different combustion characteristics, and sludge samples from different sources perform similarly but also have some different points.

Fitted curves by linear calculation and actual curves of blends were compared to research on the interaction between sludge and coal in the co-combustion process. The method can also be applied to study the interaction between two, or several, other substances in a certain temperature-programmed process.

The differences between actual and fitted DTG and DSC curves show the same results. The interaction between coal and sludge mainly takes place during the devolatilization and combustion period. Sludge can catalyze and promote the combustion of coal, which is caused by the large amount of inorganic salts contained in the sludge.

The tests under different heating rates show that the interaction between coal and sludge has a higher intensity when heating rate increases. Therefore, the interaction between the two fuels cannot

be neglected in industry applications. For the aspect of heat release, co-combustion of coal and sludge is more efficient than single burning.

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