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Oxy-fuel combustion characteristics and kinetic parameters of lignite coal from thermo-gravimetric data

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ABSTRACT

Combustion characteristics of Yi Min lignite and Zhun Dong lignite in O_2/CO_2 atmosphere were investigated by thermo-gravimetric analysis. The effects of the coal type and O_2 concentration were studied. The results indicate that the increasing of O_2 concentration can significantly improve the combustion performance especially when the O_2 concentration is less than 60%. The activation energy corresponding to the YM lignite combustion in O_2/N_2 and O_2/CO_2 atmospheres was evaluated by two isoconversional methods. The calculation results showed that further research is required whether the activation energy calculation with isoconversional methods for complex reaction process can be used as a basis for judging the degree of the reaction difficulty.

1. Introduction

Human-induced warming of the climate system has been accepted as one of the most important problems driving climate change and needs to be controlled. Most of the observed global warming over the last 50 years was likely caused by greenhouse gas forcing, with the dominant contributor being CO_2. Fossil fuel combustion systems such as coal-fired power plants are considered to contribute 33–40% of all anthropogenic emissions of carbon worldwide [1].

With the increasing growth in demand for electrical power, developing countries such as China have constructed new coal-fired power plants to keep pace with economic development and demands. In China, coal is a much more abundant resource than other fossil fuels, such as oil and natural gas, so China chooses coal as the primary fuel for power production. Therefore, we must reduce the CO_2 emission of coal-fired power plants and try to maintain the position of coal as a viable energy option in a carbon-constrained world. There are several potential strategies to achieve this goal: (1) improving the efficiency of power plants; (2) introducing of combined cycles-as-fired or IGCC, which can reach high thermal efficiencies; (3) replacing hydrocarbon fuels with renewable resources; (4) capturing and storing CO_2 from conventional plants [2]. Currently, capture and storage of CO_2 has attracted attention from the worldwide scientific community. Carbon capture technologies can be generally divided into three main categories: post-combustion capture, pre-combustion capture and oxy-fuel combustion capture. Compared to other technologies, oxy-fuel combustion is less expensive and more effective according to several techno-economic assessment studies [3,4]. In oxy-fuel combusiton technology N_2 is replaced with CO_2 in order to obtain a high CO_2 concentration in the flue gas. In most concepts, oxy-fuel combustion technology uses recycled flue gas to lower the flame temperature. To implement this technology, significant fundamental research should be done on various aspects of the system, including heat and mass transfer effects, the process and kinetics of combustion, pollutants emissions and ash deposition chemistry. Tan et al. [5] have performed oxy-fuel combustion experiments using a variety of coals. Their results showed that oxy-fuel combustion can be used to retrofit existing coal-fired power plants or to build new power plants with zero emission potential. Shaddix and Molina [6] conducted experiments in a combustion-driven laminar flow reactor that revealed that the use of oxy-fuel recycle combustion can produce ignition times and volatile flames similar to those of air–coal combustion. The successful research results mentioned above indicate that oxy-fuel combustion is possible and merits further investigation. Recently, several extremely large reserves of lignite have been discovered in the mid-west of China, it is foreseeable that many new lignite-fired power plants will be built in the near future. Therefore, it is meaningful to do the research of the oxy-fuel combustion with the lignite coal. This research may contribute to the CO_2 emission reduction.

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Thermo-gravimetric (TG) study is a traditional combustion analysis method and has been widely used for the assessment of the combustion behavior of coal. TG study is already being used in research on oxy-fuel combustion. Niu et al. [7] studied three types of pulverized coal in the oxy-fuel atmosphere by thermo-gravimetric analysis and suggested that the O_2 concentration should be less than 40% in the oxy-fuel combustion. The maximum O_2 concentration Niu chose was 60%. Li et al. [8] finished TG analysis with one coal type and four different O_2 concentrations. Their results showed that the burning process of pulverized coal in an O_2/CO_2 environment is delayed when compared with that in an O_2/N_2 environment at the same O_2 concentration. When the O_2 concentration increases up to a maximum of 80%, the combustion rate increases and the burnout time is shortened.

In this study, two different kinds of lignite coal and five O_2 concentrations ranging from 21% to 100% were chosen to perform TG experiments. The oxy-fuel combustion process can be investigated more comprehensively with the corresponding combustion characteristic parameters from the TG–DTG curves. Furthermore, kinetic analysis of combustion by a model-free isoconversional method was used to obtain data on the reaction kinetics of oxy-coal combustion with Yi Min lignite in different O_2 concentration atmospheres.

2. Experimental

2.1. Lignite coal sample

Two kinds of representative Chinese coal were selected from two different lignite coal mines. These samples were denominated according to the location of the coal mines: YM lignite from the Yi Min coal mine; and ZD lignite coal from the Zhun Dong coal mine. The proximate and ultimate analyses data of these coal samples were listed in Table 1. The coal samples were first crushed and pulverized with a bench-scale mill in the laboratory, and then sieved through 200-mesh and 325-mesh screens to obtain a sample size between 45 μm and 75 μm. Finally, the samples were stored in sealed sample bags for TG experiments.

2.2. Apparatus and procedure

TG experiments were performed in a TGA/SDTA851e thermo-gravimetric analyzer produced by Mettler-Toledo International Inc. The mass loss signal (TG) and mass loss rate signal (DTG) were recorded continuously under a stable heating rate. Before the formal tests started, we performed the preliminary experiments three times to examine the reproducibility of the experimental conditions. One coal type (YM) and one test condition (O_2/N_2 21%) were selected for the preliminary experiments. The TG curves perfectly overlapped and the error was acceptable.

In this paper, first we compared the two kinds of lignite coal with their oxy-fuel combustion characteristics data. Both the YM lignite and ZD lignite were used for the TG experiments. Approximately 10 mg of pulverized lignite coal sample was heated at a linear rate of 30 K/min. The temperature was raised from 298 K to 1073 K to ensure that the weight curve was leveled and stable. The combustion atmospheres included O_2/N_2 and O_2/CO_2 with an O_2 concentration of 21%, and O_2/CO_2 with O_2 concentrations of 40%, 60%, 80% and 100%. All gas sample purities were over 99.9%, and N_2 purity was over 99.999%. Two mass flow meters were used to control the O_2 concentration and the total gas flow was stable at 100 ml/min.

After the comparison of the oxy-fuel combustion characteristics data with the two lignite coal, YM lignite was selected to do the kinetics analysis in different oxygen concentration atmospheres. According to the recommendations for performing kinetic computations on thermal analysis data from the ICTAC Kinetics Committee [9], three different heating rates were applied in each atmospheres (10 K/min, 30 K/min, 50 K/min). Furthermore, in order to demonstrate that there was no sample mass dependence, three runs were performed on samples of three markedly different masses (5, 10 and 15 mg) in 100% concentration N_2 atmosphere. The three TG curves were identical within the experimental error.

3. Results and discussion

3.1. Effects of O_2/CO_2 atmosphere compared to O_2/N_2 using TG analysis

Molina and Shaddix [10] conducted ignition experiments in a laminar flow reactor using pulverized, highly volatile bituminous coal. Their data showed that the exchange of CO_2 for N_2 did not significantly affect the devolatilization process. Kiga and colleagues [11,12] found that the high heat capacity of CO_2 contributed to the delayed flame ignition in oxy-fuel combustion. Rathnam et al. [13] measured the reactivity of four pulverized Australian coals under simulated air and oxy-fuel atmospheres with a drop tube furnace. Their results showed that all kinds of coal had higher apparent volatile yields in CO_2 than in N_2. However, burnout characteristics in the two atmospheres were different based on the coal types used. To explore and verify the effect of CO_2 on coal combustion, thermo-gravimetric experiments were performed in a 21% O_2/79% CO_2 environment and compared with experiments performed in a 21% O_2/79% N_2 environment with YM and ZD coal. The TG and DTG curves are shown in Figs. 1 and 2.

It can be seen from the curves that there are obvious differences between the two combustion processes in the two different atmospheres and for the two different lignites. The DTG curves of both the YM coal and ZD coal in an O_2/N_2 atmosphere are higher and sharper when compared with an O_2/CO_2 atmosphere. The maximum mass-loss rates of both the two lignites in an O_2/N_2 atmosphere are obviously higher than that in an O_2/CO_2 atmosphere. These phenomena indicate that because the heat capacity and transport property of the two atmospheres are different, the combusiton behavior in 21% O_2/CO_2 changed compared with 21% O_2/N_2 atmosphere. As a result, the ignition and burnout temperature in an O_2/CO_2 atmosphere are higher than that in an O_2/N_2 atmosphere. When the experiment atmosphere changed from 21% O_2/N_2 to 21% O_2/CO_2, the maximum mass-loss temperature of YM lignite increased 8.1 K and the maximum mass-loss rates decrease 0.042%/s. On the other hand, the maximum mass-loss temperature of ZD only increased 3.9 K and the maximum mass-loss rates decrease 0.027%/s. Because the highly volatile YM coal sample will generate more pores and be filled with the ambient gas during the

Table 1
Proximate and ultimate analysis of the tested coal samples.

Sample	Proximate analysis				Qb_{ad} (J/g)	Elementary analysis				
	M_{ad}%	A_{ad}%	V_{ad}%	FC_{ad}%		C_{ad}%	H_{ad}%	N_{ad}%	St_{ad}%	O_{ad}%
YM	13.88	11.01	31.95	43.16	22,319	58.28	4.69	1.1	0.18	10.86
ZD	23.45	2.94	24	49.61	21,572	57.91	2.73	1.04	0.32	11.61