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Proceedings of the Combustion Institute 36 (2017) 2147–2154

Proceedings of the Combustion Institute

www.elsevier.com/locate/proci

Kinetic study on in-situ and cooling char combustion in a two-step reaction analyzer

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Received 4 December 2015; accepted 12 July 2016
Available online 26 July 2016

Abstract

Cooling char, which is prepared in an inert atmosphere and then reacted with oxygen after cooling, has been widely used in the kinetic study of char combustion. However, there is no cooling process in commercial combustion systems and thus the impacts of the cooling process on char reactivity remain unclear. To illustrate this question, a two-step reaction analyzer was developed to study the combustion kinetics of in-situ char, which was produced at a constant high temperature in an inert atmosphere and then directly reacted with oxygen at different temperatures. Two types of Chinese coal, Shenhua sub-bituminous coal and Yangquan anthracite, were selected as samples. The results showed that the reaction rate of in-situ char was about 1.1–1.25 times of that of rapid cooling char, suggesting that the cooling process has a significant effect on char reactivity. Then the combustion rate and structure of chars cooled at different cooling rates were investigated to determine the reason for char deactivation during the cooling process. The cooling char reactivity decreased with the decreasing cooling rate, but the specific surface area did not decreased considerably. This result revealed that char deactivation during the cooling process was independent of specific surface area. As the cooling rate decreased, the oxygen chemisorption capacity (active surface area) of cooling char decreased for Shenhua coal, indicating that the observed char deactivation during the cooling process might be caused by the decrease of active surface area. In addition, Yangquan anthracite, as a high rank coal, was less sensitive to the cooling process due to its higher carbon crystallites and provision of fewer active sites for oxidation.

Keywords: Combustion kinetics; In-situ char; Two-step; Micro-fluidized bed; Cooling

1. Introduction

The coal combustion process consists of the initial devolatilization and the later combustion of the produced char. The second step plays an important role and the char burnout time takes about 70–80% of the particle residence time in actual combustion

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http://dx.doi.org/10.1016/j.proci.2016.07.045
1540-7489

Table 1
Proximate and ultimate analyses of coal samples.

Samples	Proximate analysis (wt%, dry basis)			Ultimate analysis (wt%, dry basis)				
	Volatile	Ash	Fixed carbon	C	H	O[a]	N	S
YQ	12.04	15.02	72.94	73.04	3.28	7.32	0.92	0.42
SH	33.72	7.87	58.41	70.58	4.10	13.85	0.85	2.75

[a] By difference.

systems [1]. Therefore, a better understanding of char combustion kinetics is essential to the design and operation of commercial coal combustion systems.

Over the past decades, extensive research has been conducted on char combustion kinetics [1–7]. Baum and Street [2] described the char oxidation rate based on the apparent activation energy. Smith [4] proposed a fundamental intrinsic reaction model and defined an intrinsic reaction rate coefficient. Hurt et al. [5] proposed a carbon burnout kinetic model, which incorporates the effects of thermal annealing and ash inhibition. All these reaction models have been applied by CFD to predict the char combustion rate in commercial coal combustion systems, which requires kinetic parameters including activation energy and pre-exponential factor. Therefore, the method to obtain these kinetic parameters becomes a particularly important issue. Summarizing the previous studies on measuring kinetic parameters, the chars were first produced in an oxygen-deficient atmosphere and then cooled down rapidly. The combustion kinetic parameters of the produced char were then measured in another reactor such as the thermal gravimetric analyzer. An additional cooling process took place between the two steps. However, in commercial coal combustion systems, after devolatilization the produced char reacts with oxygen directly without any cooling process. So it is necessary to clarify the effects of the cooling process on char combustion rate.

Che et al. [8] investigated the impacts of the cooling and reheating process on the transformation of N from char-N to NO or N_2O. Results indicated that the cooling and reheating process brought about a reduction of NO emission during char combustion [8]. Kasaoka et al. [9] found that when char was cooled from 600 °C to 100 °C after devolatilization, there was no change in char reactivity. However, the devolatilization temperature was too low and char was insufficiently devolatilized. Xu et al. [10] used a micro fluidized bed to study gasification kinetics of coal char that was prepared in CO_2 and directly gasified at the same temperature. Results indicated that the annealing (cooling) during devolatilization had great impacts on the gasification kinetics and behavior of char [10]. However, the char used in this study was produced at different temperatures and would be annealed to different extents, as suggested by Jensen et al. [11]. The different devolatilization temperatures are likely to affect char reactivity. Therefore, the influence of the cooling process on char combustion kinetics has not been clearly clarified as it is difficult to avoid the effects of different devolatilization temperatures and atmospheres on char reactivity. The kinetic study of in-situ char, which was prepared at a constant temperature in an inert atmosphere and then combusted at different temperatures, has not been achieved until now. The major problem is how to ensure the appropriate in-situ test conditions of char combustion kinetics.

We developed a two-step reaction analyzer to study combustion kinetics of in-situ char. Coal was rapidly devolatilized at a constant temperature in a laminar flow reactor. The produced char with high temperature was then directly combusted in a micro fluidized bed that was preset at a lower temperature.

Based on the developed two-step reaction analyzer, this paper focuses on the combustion kinetics of in-situ char. The reaction mechanism and kinetic parameters between in-situ char and rapid cooling char were compared. The combustion kinetics of chars prepared at three different cooling rates were analyzed to further clarify the influence of the cooling process on char structure and reactivity. Finally, the chemical composition, specific surface area and chemisorption capacity of three cooling chars were compared.

2. Experiment

2.1. Materials

Two different kinds of coal were selected as samples in this study: Yangquan anthracite (YQ) and Shenhua sub-bituminous coal (SH). Samples were ground and sieved to a diameter between 74 and 100 μm to minimize pore diffusion. Prior to experiments, samples were dried at 105 °C for 2 h. Their proximate and ultimate analyses are given in Table 1.

2.2. The two-step reaction analyzer

Figure 1 shows the schematic diagram of the two-step reaction analyzer that enables the kinetic study of in-situ char combustion. The analyzer consists of three parts: the laminar flow reactor where rapid devolatilization occurs, the micro