Fundamental Reaction Characteristics of Pulverized Coal at High Temperature

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In order to enhance the overall thermal efficiency in blast furnace system as well as electric power generation systems by coal combustion/gasification, it is necessary to elucidate fundamental reaction characteristics of pulverized coal at high temperature. The experiments were carried out using a horizontal pulverized coal reactor with a pre-combustor to produce vitiated air at high temperature and high-oxygen concentration. The reaction behavior of coal during combustion and/or gasification are elucidated by means of sampling and analyzing both reacting particles and reaction gases as well as optical measurement of the instantaneous particle temperature. Evolution rate of the volatile matter for various types of coal at high temperature is obtained from the Arrhenius plots.

As a result, the volatile matter is rapidly evolved as soon as coal particles are introduced into the furnace since temperature of the vitiated air increases sufficiently. Result shows that the evolution rate of volatile matter does not depend on coal types. Activation energy and frequency factor of the evolution rate of volatile matter for various types of coal remain almost constant in the particle temperature range of 2200 to 2700 K, even when other experimental conditions were varied. In the char combustion region, char structure was found to affect the reaction rate of fixed carbon. It is observed that coal, which forms a network-type of char, indicated reaction rate of fixed carbon which did not always increase even when the vitiated air temperature was high enough. For a balloon-type char, on the other hand, the reaction rate of fixed carbon decreased with an increase of the vitiated air temperature.

KEY WORDS: blast furnace; ironmaking processing; coal and coke.

1. Introduction

In order to enhance the overall thermal efficiency of blast furnace system as well as electric power generation systems by coal combustion/gasification, advanced coal reaction technologies at high temperature have been developed worldwide. Recently, pulverized coal combustion technologies at high-temperature and high-oxygen concentration have been applied to not only integrated gasification combined cycles (IGCC), but also pulverized coal injection (PCI) into the blast furnaces. In order to design the such systems, it is necessary to study quantitatively the characteristics of coal reaction at high temperature, the evolution rate of the volatile matter, conversion of the fixed carbon, detail reaction behaviors and the like.

Many studies on pulverized coal combustion/gasification have already been reported by a number of researchers. Young and Niksa\(^1\) studied the combustion rates for selected low-rank coal chars, Baum and Street\(^2\) tried to formulate the combustion behavior of coal particles while Smith \textit{et al.}\(^3\) summarized current detailed coal reaction characteristics. However, quantitative characteristics on coal combustion at high temperature exceeding 2000 K, for example the detail reaction behaviors, the evolution rate of volatile matter and so forth, have not been always studied enough. For instance, the outline of high rate PCI operation in Japan was reported by Shimizu \textit{et al.}\(^4\) This technology has been developed in order to enhance the productivity of iron and to reduce fuel cost owed to decreasing the coke consumption. Inaba\(^5\) studied the relationship between the rate of pulverized coal injection and the combustion efficiency in the tuyere. Artyama \textit{et al.}\(^6\) studied movement of pulverized coal particles, based on the direct observation. Shen \textit{et al.}\(^7\) tried to improve pulverized coal combustibility by addition of KMnO\(_4\) as a catalyst. Jamaluddin \textit{et al.}\(^8\) and Tsuge \textit{et al.}\(^9\) tried to develop a mathematical model of the pulverized coal combustion in the blowpipe and raceway. The effect of volatile matter and oxygen enrichment on the combustion efficiency was reported by Iwanaga \textit{et al.}\(^10\).

However, precise and quantitative knowledge on the reaction behaviors at high temperature such as more than 2000 K of particle temperature have not been elucidated yet. For the development of such advanced coal reaction technologies, therefore, it is desirable to obtain or measure key parameters like the evolution rate of volatile matter, reaction rate of fixed carbon, changes of the specific surface area and particle temperature.

In this study, pulverized coal combustion/gasification at high temperature are performed by using a horizontal pulverized coal reactor with a pre-combustor to produce vitiated-
ed air of high-temperature and high-oxygen concentration. The fundamental reaction parameters like the evolution rate of volatile matter, the reaction behavior of fixed carbon and morphological change of char particles are elucidated by means of sampling and subsequently analyzing the reaction gas and reacting particles as well as measuring the instantaneous particle temperature during the reaction. Based on the results obtained, the evolution rates of volatile matter for various types of coal are shown as an Arrhenius plot. The relationship between the conversion of fixed carbon and the char morphology is also discussed.

2. Experimental Procedures

Figure 1 shows a schematic diagram of a horizontal pulverized coal reactor with a pre-combustor employed in this study. The dimensions of the furnace are 2.1 m long and 0.12 m in inner diameter. In the pre-combustor, high-temperature vitiated air is produced by combustion of natural gas with oxygen-enriched ambient air. The main coal reactor consists of two parts, namely; the front part is cooled by water since the particle temperature may exceed 2000 K and the rear part which is insulated by refractory material to reduce the heat loss. The front part of main reactor is designed with four sampling ports while the rear part is designed with six sampling ports, all at a distance of 0.15 m between them. The gas velocities at the inlet of pulverized coal and at the main furnace are about 20 m/s and about 10 m/s, respectively. Therefore, the residence time in average of the main furnace has about 0.01 s.

Five types of coal with differing properties are selected as samples for the present experiments. Properties of the coal types tested are shown in Table 1, where fuel ratio denotes content ratio of fixed carbon to volatile matter in coal. As seen from the table, the content of volatile matter for C coal is the lowest of the five. The coal types tested were pulverized with 80% of mass fraction less than 72 μm. Table 2 shows the experimental conditions, where heat load is fixed at a value of 44 kW.

In the experiments, both burning particles and reaction gases were sampled at each sampling port by the water-cooled probe, at the tip of which nitrogen gas was injected for dilution. From the sampled particles, volatile matter and fixed carbon contents were analyzed by a thermobalance. Char structure of the reacting particles was observed using an optical microscope. Specific surface area of the reacting particles was also measured by BET (Brunauer–Emmett–Teller) method. Particle temperature was optically measured using an instantaneous two-color pyrometer along the furnace axis.

3. Results and Discussions

From the experiments carried out, the behavior of coal types on reaction zone, vitiated air conditions and their influences on the reaction and the evolution rate of volatile matter could be observed. These are discussed in the following subsection.

3.1. Effect of Coal Types on Reaction Behaviors

Figure 2 shows fundamental profiles of gas species concentration and particle temperature along the furnace axis for A coal. The stoichiometric ratio, vitiated air temperature and oxygen concentration are maintained at 1.2, 1300 K and 21%, respectively, as indicated in the figure. From the figure, the maximum particle temperature exceeds 2500 K near the coal injection point, at which the volatile matter burns rapidly. At the same time as O2 is consumed, CO2 and...
CO are produced. The profiles of gas species concentration and particle temperature along the furnace axis of other coal types that almost similar.

Figures 3 and 4 show the effect of vitiated temperature on the reaction behavior for A and C coals, respectively. These figures show profiles of conversions of volatile matter (VM) and fixed carbon (FC) as well as specific surface area (SSA) for reacting particles sampled. The vitiated air temperature was varied from 1 300 to 1 500 K. These figures show that volatile matter evolved rapidly as soon as the coal is supplied into the main reactor since the vitiated air temperature is high enough. Evolution behavior of volatile matter increases with an increase of the vitiated air temperature. This characteristic emphasizes the importance of temperate on the volatile matter evolution.

Comparing Fig. 3 and Fig. 4, the evolution tendency of volatile matter for C coal shows a little bit slower rates than that of A coal. This is due to the difference of fuel ratio as indicated in Table 1. For A coal, the conversion of fixed carbon does not depend on the vitiated air temperature. The C coal, on the other hand, conversion of fixed carbon decreases with an increase of the vitiated air temperature. This may be due to the difference in char structure since the profile of specific surface area of A differs from that of C coal. The specific surface area for the case of A coal increases with an increase in the vitiated air temperature. However, for C coal, converse is observed, while the maximum specific surface area for C coal is almost half of that for A coal.

In order to clarify the above observation, char structure of the reacting particles for each coal type was observed by using an optical microscope. Figure 5 shows the cross-section of char particles for A and C coal types. From the photographs, the char particles for A coal are classified as a network-type. C type of coal, on the other hand can be classified as a balloon-type char formed during the reaction. Generally, the char particles, forming a network-type, have a large specific surface area since it is characterized by a lot of fine pores inside the particle. However, the interior balloon type of char much more empty. The observed results therefore, can well explain the tendencies of specific surface area appearing in Figs. 3 and 4.
3.2. Effect of Vitiated Air Conditions on Reaction Behaviors

In order to elucidate the effect of change of coal feed rate into a blast furnace on reaction behaviors by means of the lab-scale experimental furnace, stoichiometric O₂ ratio have to controlled at the same coal feed rate although the flow behavior in the furnace show a slightly changes. Profiles of the gas species concentration and the conversion of fixed carbon against the furnace axis for B coal is shown in Fig. 6. For the experimental condition of O₂ stoichiometric ratio of 0.8, corresponding to the condition of high coal feed rate in practical blast furnaces. Comparing the upper figure (O₂ ratio=1.2) and the lower figure (O₂ ratio=0.8), the fixed carbon consumption is reduced thus increasing CO concentration with a decrease of O₂ ratio. CO generation under the reducing condition is mainly caused by solution loss reaction. The reason why the tendency of O₂ concentration does not always harmonize with that of fixed carbon conversion is caused by non-plug flow in the furnace. Furthermore, the concentration obtained by gas sampling only shows a cup-mixing concentration.

From the figure, a large amount of unburned char remains in the particles when the O₂ ratio is low. Therefore, it is necessary to enhance the conversion of fixed carbon, which may be attained by oxygen enrichment. Figure 7 shows the profiles of the gas species concentration and the conversion of fixed carbon for B coal. The experimental conditions are indicated in the figure. Comparing the results shown in the Fig. 7 with O₂ ratio 0.8 than that of Fig. 6, the fixed carbon conversion and CO concentration increase as a result of O₂ enrichment. This is caused by the enhancement of solution loss reaction. In order to check the morphological change during the reaction, specific surface area was measured for each sample. Figure 8 shows profiles of specific surface area of the reacting particles at 21 and 31% of oxygen concentration in the vitiated air. It can be seen from the figure that the specific surface area of reacting particles increases with an increase of the oxygen concentration. This result suggests that the method of O₂ enrichment contribute to the enhancement of the char reactivity due to the solution reaction by CO₂ and H₂O.

3.3. Evolution Rate of Volatile Matter

In this experiment, the instantaneous particle temperature is optically measured, and the conversion of volatile matter in the reacting particle analyzed. By this way it is possible to estimate the evolution rate of volatile matter based on the particle temperature. However, the evolution rate was rapid, as already presented in Figs. 3 and 4, that the constant rate model could be adopted in this study. The evolution rate of volatile matter from the coal particles can be expressed as in Eq. (1).

\[
\frac{dX_{VM}}{dt} = kA \tag{1}
\]

where, \(X_{VM}\) is the conversion of volatile matter, \(t\) is the residence time of coal particle in the main furnace, \(A\) indicates the specific surface area of raw coal particles and \(k\) is the rate constant of volatile matter evolution. The reason why the constant model was adopted to reveal the evolution rate of volatile matter as follows. As shown in Figs. 3 and 4, the
volatile matter was rapidly evolved as soon as the coal particles are injected, and volatile matter evolution almost completed for a few moments. It is because the particle temperature goes up instantaneously. This experimental results support that the evolution phenomenon does not depend on either the infinitive evolution fraction of volatile matter or the initial specific surface area. The rate constant of volatile matter evolution is correlated to the Arrhenius type equation based on the particle temperature \( T_p \) as indicated in Eq. (2).

\[
k = k_0 \exp \left( - \frac{E}{RT_p} \right)
\]

where \( k_0 \) and \( E \) are the frequency factor and the activation energy, respectively. \( T_p \) is measured at the first sampling port by using a two-color pyrometer. The residence time from the injection point until the first port is about 7.5 ms.

In order to compare the rate constant of this study with the previous results, \(^1\) data were fitted with the commonly used first order model as shown in Fig. 9. From the result of previous studied shown that under condition of temperature less than 2000 K, the rate constant differs according to the types of coal and combustion condition. On the other hand, in our experiment for particle temperature exceed 2000 K shown that the evolution rate of volatile matter was not significant increased and kept nearly constant, even if the combustion condition were varied.

Based on the obtained results suggest that the evolution rate of volatile matter does not depend on coal types or the experimental conditions since the particle temperature is high enough. It is evident that the volatile matter evolution is generally controlled by heat transfer characteristics in the furnace. It is therefore, in this study tried that the rate constant was normalized by specific surface area of coal particles. Figure 10 shows the Arrhenius plots of rate constant of volatile matter evolution for various coal types. From the figure shows that in the range of particle temperature from 2200 to 2700 K, the activation energy and the frequency factor for the evolution rate of volatile matter is about 41.4 kJ/mol and about 148.4 s\(^{-1}\), respectively.

4. Conclusion

Fundamental reaction characteristics of pulverized coal under high temperature condition have been studied by using a horizontal pulverized coal reactor with a pre-combustor. Five types of coal were used so as to elucidate the specific influences of the major parameters. From the obtained results and foregoing discussion, the following are the conclusions:

(1) The evolution rate of volatile matter increases with an increase of the vitiated air temperature. This behavior is the same regardless of the type of coal used.

(2) Fixed carbon conversion depends on the char type. For the coal type, which form a network-type char, an increase of the vitiated air temperature does not always affect the conversion of fixed carbon. For a balloon-type char, on the other hand, the fixed carbon conversion decreases with an increase of the vitiated air temperature.

(3) Under the weak reducing conditions, fixed carbon conversion decreases since the stoichiometric oxygen ratio is less than unity. However, the conversion of fixed carbon can be enhanced by means of oxygen enrichment in the vitiated air.

(4) The evolution rate of volatile matter does not depend on coal types. However, it is depend on the particle temperature.

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