Korean J. Chem. Eng., **30**(5), 1034-1038 (2013)
DOI: 10.1007/s11814-013-0018-7

A comparison of spontaneous combustion susceptibility of coal according to its rank

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(*Received 19 November 2012 • accepted 6 February 2013*)

Abstract−This study investigated spontaneous combustion susceptibility of coal according to the rank. To estimate the spontaneous combustion susceptibility of coal, both crossing-point temperature (CPT) measurement and gas analysis by using gas chromatography (GC) were performed. For the experiment, Eco coal and Kideco coal, Indonesian lignite, and Shenhua coal that is Chinese bituminous coal were used. The lignite such as Eco coal and Kideco coal contains more functional groups that easily react to oxygen more so than Shenhua coal. For this reason, the lignite is more easily oxidized than bituminous coal at low temperature, which results in high O_2 consumption, increase in CO and CO_2 generation, and low CPT. Although the CPT of Eco coal and Kideco coal is identical to each other as they are the lignite, Kideco coal has a lower initial oxidation temperature (IOT) and maximum oxidation temperature (MOT) than those of Eco coal. This means that although each coal has the same rank and CPT, spontaneous combustion susceptibility of coal may vary because the initial temperature of the coal at which oxidation begins may be different due to the substances that participate in oxidation.

Key words: Coal Rank, Crossing-point Temperature, Spontaneous Combustion, Low Temperature Oxidation

INTRODUCTION

The spontaneous combustion susceptibility of coal causes difficulties in transporting, storing, and using coal, which is critical in terms of safety and economy [1,2]. Several methods are available to determine the spontaneous combustion susceptibility of coal, such as adiabatic oxidation [3-5]; high temperature activation energy [6, 7]; crossing-point temperature [8-12]; and gas analysis methods [13] If air or oxygen is supplied in the oven that contains the coal by increasing the temperature inside the oven, the temperature of the coal will increase along with the temperature of the oven, and at a certain point, the coal temperature will exceed the oven temperature due to self-heating through the oxidation of coal. This point at which the coal temperature crosses that of the oven is called the crossing-point temperature (CPT). Research on CPT measurement was first done by Nubling, et al. [14]. CPT measurement has been used for evaluating spontaneous combustion susceptibility of coal. CPT measurement in previous studies was conducted according to coal properties such as rank, particle size, moisture content, and application conditions such as the flow rate of oxygen, heating rate, and the amount of coal samples [9-12]. Since the CPT measurement method measures the temperature of part of all coal contained in the vessel, it may decrease the reliability of the results due to the property of coal that is not a single substance.

Gas analysis using gas chromatography (GC) is used to estimate spontaneous combustion susceptibility of coal through analyzing the concentration of CO and CO_2 that is generated when coal reacts with oxygen and the oxygen consumption [15,16]. In general, the higher the spontaneous combustion susceptibility of coal, the more the O_2 is consumed and the more CO and CO_2 generate even at low temperature [17-19].

By combining the CPT measurement and GC analysis method, we can analyze the spontaneous combustion susceptibility of coal and complement any shortcomings that only we can measure the part of coal temperature during CPT measurement.

In this study, we measured CPT of two kinds of lignite and one kind of bituminous coal in order to estimate the spontaneous combustion susceptibility of coal according to rank. In the process, we analyzed the CO and CO_2 concentration that generated in the process by using the GC. During the experiments, we fixed the factors that may affect the result of CPT measurement: gas flow rate, heating rate, moisture content, and size of the coal.

EXPERIMENTAL

1. Coal Sample

We used Eco coal and Kideco coal, which are Indonesian lignite, and Shenhua coal, that is, Chinese bituminous coal. Among the coals, we selected lump coal that was more than 10 cm in diameter for the samples to minimize the effect of weathering. The lump coal was crushed and sieved in order to obtain particles that were 180-425 μm in size for use. To remove any effect caused by moisture during the CPT measurement, the coal was dried at 107 °C under a nitrogen atmosphere for 12 hours.

2. Experimental Apparatus and Procedure

A conceptual diagram of the CPT measurement apparatus used in the experiment is shown in Fig. 1. Two vessels that contain a coal sample are located inside an oven in which the temperature is programmable. To each vessel, nitrogen or air is selectively supplied by manipulating the valve. The gas is supplied to the vessel by controlling the flow with a mass flow controller (MFC), and when it is passing through the copper tube coil inside the oven, the temperature of the gas is adjusted to the oven temperature. The gases that

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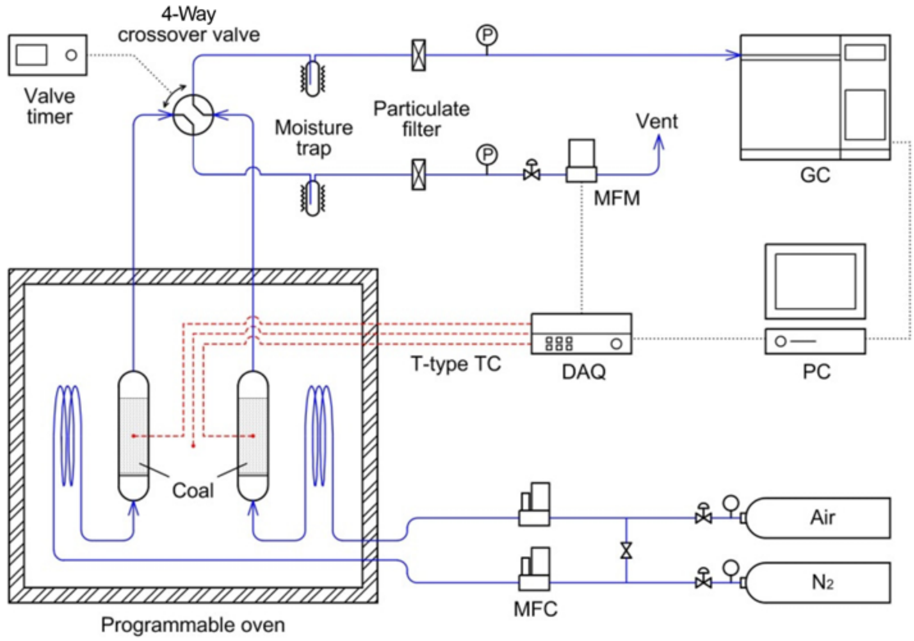

Fig. 1. Schematic diagram of experimental apparatus.

pass through the vessels are sent to the GC (6890N, Agilent Technologies) for measuring components, or the mass flow meter (MFM) for measuring the flow rate after reaction, according to the selected 4-way crossover valve location that changes periodically. The moisture trap and particulate filter are attached on the front area of the GC and the MFM. The O_2, CO, and CO_2 gases emitted from each vessel were measured in the GC to which the thermal conductivity detector and two columns (60/80 Molecular Sieve 13X, 80/100 Porapak N, SUPELCO) are installed.

We loaded 35 g of the coal sample dried in each vessel and installed them inside the oven that was set at 40 °C. Then, we supplied nitrogen gas of 75 mL/min for two hours in order to cause the temperature to achieve equilibrium. When the temperature of the coal inside the vessel was 40 °C, we supplied nitrogen gas to one vessel of 75 mL/min and the air to another vessel with the same flow rate and increased the oven temperature to 180 °C by 0.5 °C/min.

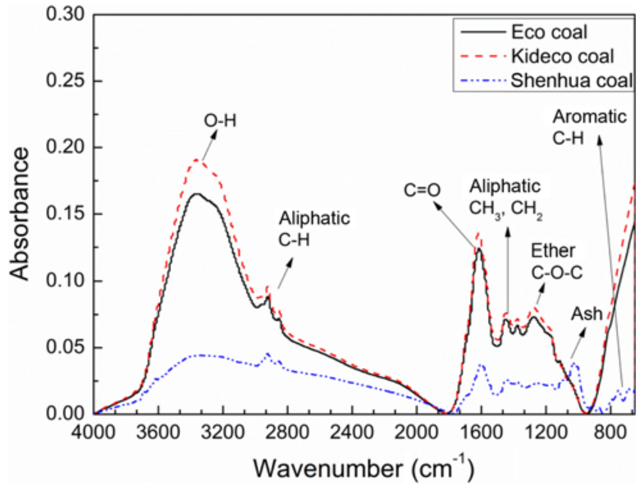

Fig. 2. FTIR spectra of coal samples.

RESULTS AND DISCUSSION

1. Properties of Samples

Table 1 shows the results of proximate analysis and calorific value analysis of the coal samples. The moisture content of the two lignite coal, Eco coal and Kideco coal, was 29.8 and 32.2 wt%, respectively. The Shenhua coal, the bituminous coal, was 8.9 wt%. The content of volatile matter in Eco coal and Kideco coal was higher than that of fixed carbon. However, the content of fixed carbon in Shenhua coal was higher than that of the volatile matter; the fixed carbon showed a higher amount than volatile matter. As for the ashes, Shenhua coal contained 14 wt% of ash content and it was higher than Eco coal and Kideco coal, which contained 2 wt%. It has been known that the lower the rank is, the more the coal will have high moisture content and the higher the volatile matter compared to the

Table 1. Proximate and calorific value analyses of coal samples

Sample	Proximate analysis (wt%) (as received basis)				Calorific value (kcal/kg) (air dry basis)
	M	VM	Ash	FC	
Eco coal	29.86	41.00	2.20	26.94	5,966
Kideco coal	32.23	40.78	2.32	24.68	6,049
Shenhua coal	8.93	30.90	14.61	45.56	6,223

fixed carbon. Coal samples used in the experiment showed such properties well.

As a result of analyzing the calorific value of the dried coal sample, the heating value of Eco coal and Kideco coal was 5,966 kcal/kg and 6,049 kcal/kg, respectively, and that of Shenhua coal was 6,223 kcal/kg. The heating value of Shenhua coal, that is, the bituminous coal, showed no notable difference from that of the lignite because Shenhua coal has high ash content per unit weight.

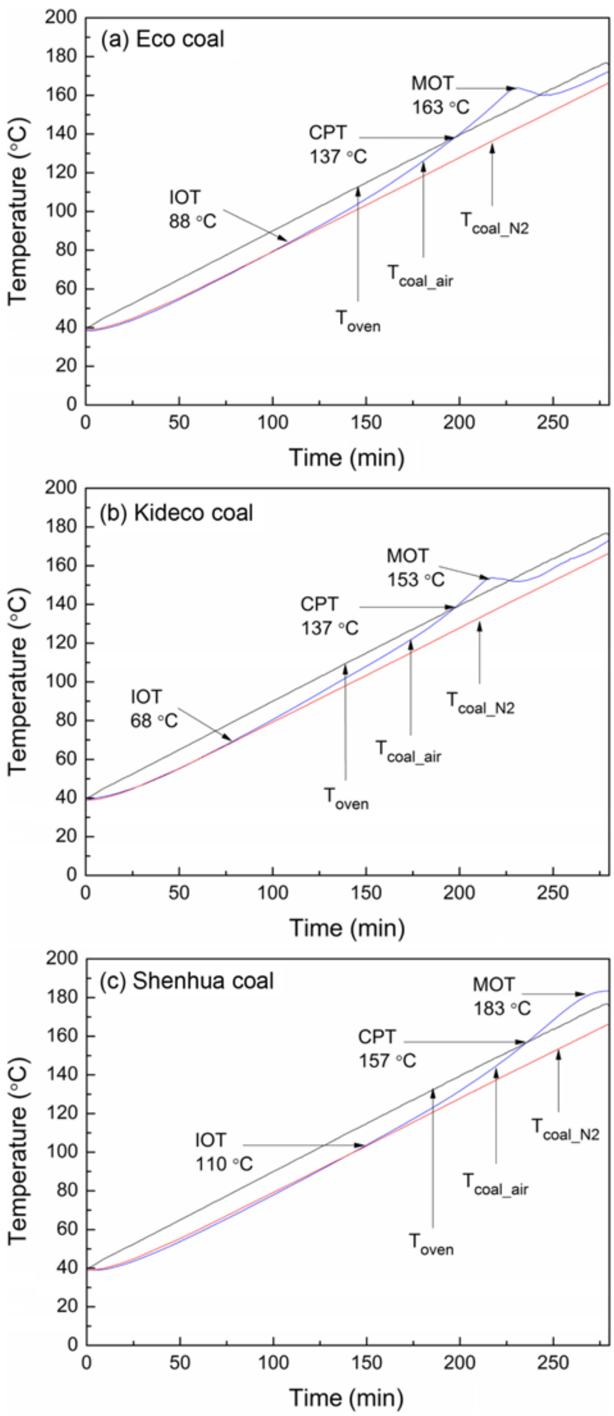

Fig. 3. Temperature profiles of air supplied coal (T_{coal_air}), N_2 supplied coal (T_{coal_N2}) and oven (T_{oven}) in CPT measurement: (a) Eco coal, (b) Kideco coal, (c) Shenhua coal.

2. FTIR Analysis

Fig. 2 shows the FTIR analysis results of coal samples. The spectra of Eco coal and Kideco coal show almost identical aspects. The broad O-H stretching peak at 3,600-3,400 cm^{-1}, C-O stretching peak at 1,700 cm^{-1}, aliphatic CH_2 and CH_3 peak at 1,450 cm^{-1} and 135 cm^{-1}, and C-O-C stretching peak at 1,300-1,100 cm^{-1} showed a high value. This explains the high moisture content and the volatile matter content shown in Table 1.

Compared to the lignite, the Shenhua coal had a low O-H peak at 3,600-3,400 cm^{-1} and a C-O peak at 1,700 cm^{-1}. However, it had an aromatic C-H peak at 900-700 cm^{-1}, which was not shown in the lignite coal and also had a mineral peak at 1,000 cm^{-1}.

3. CPT Measurement

Fig. 3 illustrates CPT measurement results of coal samples. In the figure, the temperature of the coal to which air was supplied (T_{coal_air}) is shown with the temperature of nitrogen-supplied coal (T_{coal_N2}) and the temperature of the oven (T_{oven}). The T_{coal_N2} increased in parallel with the T_{oven} till the measurement was done while maintaining a lower value than the T_{oven}. This means that there was no reaction of N_2 gas with the coal and there was no effect on the temperature of the coal. However, as for T_{coal_air}, it began to have a higher value than T_{oven} beyond a certain point. It was because coal reacted with the oxygen in the air, generated heat, and increased the temperature of the coal [9,10]. The CPT of the Eco coal and Kideco coal was the same as 137 °C. On the other hand, the CPT of Shenhua coal was 157 °C, which was higher by 20 °C than that of the lignite. This means that the lignite has good reactivity with oxygen and higher spontaneous combustion susceptibility than bituminous coal [20]. Lignite has many functional groups that can react with oxygen as shown in Fig. 2 and it has high reactivity even at low temperature compared to bituminous coal.

Both T_{coal_air} and T_{coal_N2} increased at the same slope in the initial stage of reaction, and after some time passed the slope of T_{coal_air} grew larger. This is because the temperature of the coal increased while the heat generated by oxidation of coal accumulated. We call the temperature at which T_{coal_air} started to rise more than T_{coal_N2}, the initial oxidation temperature (IOT).

The IOT of Shenhua coal was 110 °C. As for the lignite, while the CPTs of Eco coal and Kideco coal were the same as 137 °C, the IOT of Eco coal was 88 °C, and that of Kideco coal was 68 °C. The results of CPTs and IOTs show that as for the bituminous coal, oxidation started at a higher temperature than the lignite did, and even if coal has the same CPT, the IOT, at which oxidation begins, may be different.

T_{coal_air} kept increasing even after reaching the CPT and then it decreased below the T_{oven} and increased again in parallel with the T_{oven}. Decrease in T_{coal_air} means that there was no more oxidation of coal and thus no more heat accumulation. When the T_{coal_air} reaches the highest, we call this temperature the maximum oxidation temperature (MOT). The MOT of Kideco coal was 153 °C, Eco coal was 163 °C, and Shenhua coal was 183 °C.

From the above results, we can infer that Kideco coal, Eco coal, and Shenhua coal react with oxygen at lower temperatures, in that order, with Kideco coal reacting at the lowest temperature of the three.

4. Gas Analysis

Fig. 4 shows CO and CO_2 concentration during CPT measure-

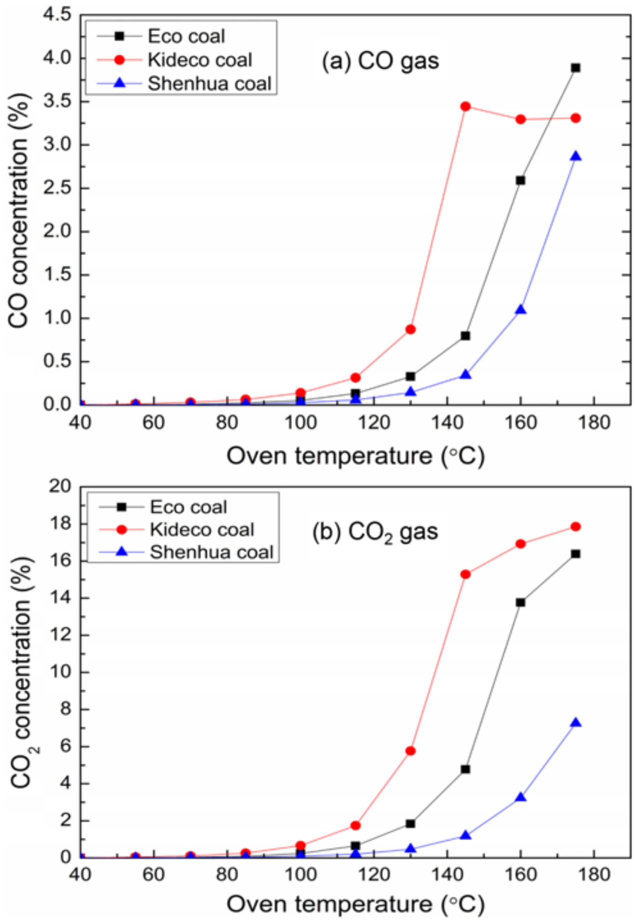

Fig. 4. Outlet gas concentration of (a) CO and (b) CO_2.

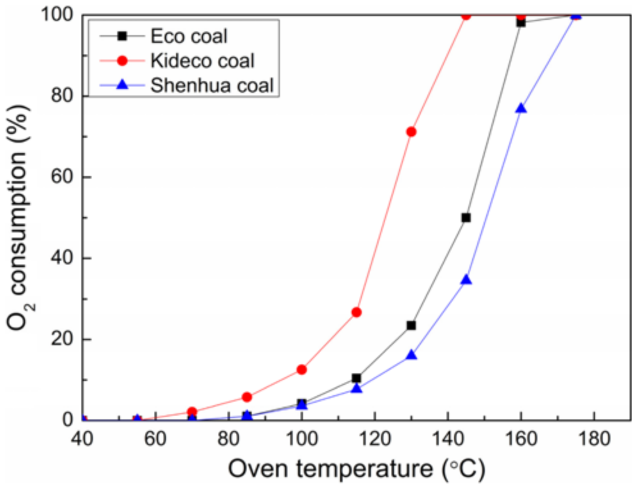

Fig. 5. O_2 consumption of coal samples.

ment, and Fig. 5 shows O_2 consumption that was calculated based on the O_2 concentrations measured at the inlet and the outlet. As a result of CO and CO_2 analysis, for all coal, the higher the temperature is, the more the gas is generated. It is due to the fact that as the temperature increased, reaction of the functional groups of the coal with oxygen rose more [21]. CO gas generation of Kideco coal achieved an equilibrium state at 145 °C and more. It is assumed to be because the supplied oxygen consumed 100% at that temperature.

Kideco coal, Eco coal, and Shenhua coal showed a lower initial temperature at which gas started to generate, in that order. This coincides with the order of IOT in CPT measurement and means that oxidation of coal began in the order of Kideco coal, Eco coal, and Shenhua coal.

Generation of both CO and CO_2 increased rapidly around the CPT. When the temperature of coal increased, decomposition occurred in a stable solid complex of coal and it produced new active sites [21]. Since new active sites enable coal to maintain continuous oxidation, the temperature of coal keeps on rising. The higher the temperature of the coal, the more the thermal decomposition increases and the generation of gases such as CO and CO_2 increases.

The O_2 consumption of Kideco coal, Eco coal, and Shenhua coal was high, in that order. It showed a similar trend to the changes in the coal temperature, CO, and CO_2 gas concentration. Kideco coal, Eco coal, and Shenhua coal had a higher temperature at which O_2 gas was consumed 100%, in that order. This means that Kideco coal, Eco coal, and Shenhua coal contain many of the functional groups of coal that participate in oxidation at low temperature, in that order.

CONCLUSIONS

This study investigated spontaneous combustion susceptibility of coal according to the rank of the coal by combining the CPT measurement and the gas analysis. The CPT of Kideco coal and Eco coal, the lignite, was the same as 137 °C and that of Shenhua coal, the bituminous coal, was 157 °C, which was higher by 20 °C than the lignite. Eco coal and Kideco coal showed a higher FTIR peak of aliphatic CH_2, CH_3 and ether C-O-C functional groups that easily react with oxygen and moreso than Shenhua coal. That is, the lower the coal rank, the more the coal contains a substance that can react with oxygen even at low temperature. Therefore, low rank coal has higher spontaneous combustion susceptibility than high rank coal.

The IOT of Kideco coal and Eco coal that had the same CPT was 68 °C and 88 °C, respectively. This means that Kideco coal started oxidation at a lower temperature than Eco coal. And, it was confirmed that in MOT, Kideco coal had a lower value than Eco coal. In the gas analysis as well, Kideco coal started to generate CO and CO_2 and consume O_2 at a lower temperature than Eco coal. This means that Kideco coal has relatively high spontaneous combustion susceptibility at low temperature, compared to Eco coal.

From this finding, we can say that although coal has the identical rank and CPT with other coal, it may have a different IOT due to the difference in substances that participate in reaction at low temperature, resulting in having different spontaneous combustion susceptibility.

Therefore, if we apply CPT measurement along with the GC analysis method, we can evaluate the spontaneous combustion susceptibility of coal more clearly. The influence of other factors, such as particle size, moisture, volatile matter, or gas flow rate, that can affect the spontaneous combustion susceptibility of coal, will be investigated using the same method.

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