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Development of Method to Evaluate the Characteristics of Coal Oxidation and Heat Generation

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Abstract

Coal reacts with atmospheric oxygen to generate heat even in general temperatures. In industries that have facilities to store coal, heat generation due to coal oxidation is recognized as a risk-causing factor in the operation. In this study, to investigate the influence of the sample mass and the thermal insulation method for the evaluation of oxidation characteristics for coals, several tests were performed. We thereby revealed that an increase in the coal sample mass produces a clear temperature rise even in oxidation performed at the same temperature. Therefore, to analyze thermal characteristics by detecting the heat of oxidation of coal at low temperature, it is necessary to use a coal sample packed bed formed with consideration given to the coal sample mass corresponding to the target process.

1. Introduction

Since Japan imports all of its coking coal, which is a raw material of coke, stable securement is an important issue. The reserves of coking coal, which can be used for coke, are approximately 10% of the total coal reserves[1] and thereby technologies to enable the use of non-caking or slightly caking coal (low-grade coal) while maintaining coke strength have been anticipated to expand resources. As a result, it is becoming possible to produce coke with stable quality even when the usage ratio of low-grade coal is increased while maintaining the strength of the coke by using the moisture control process.[2–4]

Generally, coal gradually oxidizes by taking in atmospheric oxygen and in turn generates heat.[5] Accordingly, when a large amount of moisture-controlled coal is stored in a coal bin or other similar facility, if the rate of the heat generation due to coal oxidation has been exceeding the rate of the heat release to the atmosphere for an extended period of time, the temperature of the coal in the char cistern may rapidly increase. Also, as the coal grade is lower, the heating value due to the oxidation tends to be higher.[6] That is to say, low-grade coal tends to oxidize and generate heat. Therefore, as the usage ratio of low-grade coal increases, more attention needs to be paid to safety when such coal is stored in coal bins.

In our study, the coal sample mass used was significantly varied (a few grams, a few kilograms, and a few hundred kilograms) to perform oxidation and heat generation tests to evaluate the safety of coal storage in coal bins. This report clarifies the influence of the sample mass and test conditions on the evaluation of oxidation and heat generation.

2. Main Disclosure

2.1 Experimental methods

2.1.1 Experimental samples

In this study, coal (bituminous coal including noncaking or slightly caking coal) that is generally used in the steel industry was used as experimental samples. **Table 1**[7] lists the representative coal characteristics.

2.1.2 Study using a spontaneous ignition temperature (SIT) tester

SIT testing is a standard method that can evaluate the spontaneous ignition properties of coal samples even in a small quantity.[8] As the tester, SIT-II made by Shimadzu Corporation was used.

The test procedures are as follows. One gram of coal samples for which the particle size had been adjusted equal to or smaller than 0.25 mm was prepared and put into the sample cell of the tester, then, settled in the sample holder chamber. The air inside the tester was replaced with nitrogen atmosphere. In the nitrogen atmosphere, the temperature was increased until the sample temperature reached

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110°C. After confirming that the sample temperature had reached 110°C, the temperature was lowered to the test start temperature. After it had reached the test start temperature, nitrogen inside the tester was replaced with oxygen (flow rate: 3 mL/min, hygroscopic moisture: 0%). The test was started at that point. The ambient temperature inside the tester was maintained constant throughout the test and the coal temperature was continuously measured up to three days. When the coal temperature reached 250°C in the test, the test ended. The test start temperature was in the three levels of 80°C, 90°C, and 100°C.

2.1.3 Test using a cylindrical steel can (pail)

To evaluate the influence when the coal sample mass was increased to a few kilograms, a cylindrical steel can (pail) with a diameter of 300 mm and a height of 200 mm as shown in **Fig. 1**[7] was used for testing. An insulator with a diameter of 300 mm and a height of 50 mm (made by Shin-Nippon Thermal Ceramics Corporation) was placed on the bottom of the pail. Approximately 5 kg of crushed coal samples with a mean particle size of 3 mm were charged into the insulator to a height of 100 mm. The packing density of the coal samples at that time was approximately 0.8 g/cm^3.

After charging the coal samples, a thermocouple was installed at the center at a depth of 60 mm from the surface to measure the temperature in the coal packed bed during the test. The pail containing the coal samples was settled in an injection-type thermostat. The air inside the system was replaced with nitrogen during the temperature increase from the room temperature to the test start temperature (80°C) to prevent the coal samples from oxidizing. After the temperature in the coal packed bed reached 80°C, the nitrogen in the injection-type thermostat was replaced with air (hygroscopic moisture: atmospheric humidity) at that temperature. The test was started at that point.

After the replacement with the air, the inside of the system was in natural convection where the main oxygen supply to the coal packed bed was diffused from the surface. In the test, the temperature in the coal packed bed was continuously measured up to five days. When the temperature in the coal packed bed reached 300°C or higher during the test, the test was ended. The test was performed under two different conditions: Condition 1: the ambient temperature in the injection-type thermostat was maintained constant at 80°C throughout the test; and Condition 2: considering the actual storage conditions of coal, the set temperature of the thermostat was controlled to maintain the temperature in the thermostat constant according to changes in the temperature in the coal packed bed to reduce the radiation amount in the surrounding area.

2.1.4 Evaluation test using a solid fuel storage safety tester

In a test where a large amount of coal (a few hundred kilograms)

Table 1 Characteristics of coal used[7]

Total moisture	Proximate analysis				Ultimate analysis				
	IM	ASH	VM	FC	C	H	N	S	diff. O
mass%	mass%	mass%	mass%	mass%	daf., mass%	daf., mass%	daf., mass%	daf., mass%	daf., mass%
2.4	1.8	9.1	28.6	60.5	85.8	5.1	1.9	0.7	6.5

IM: Inherent moisture, ASH: Ash contents, VM: Volatile matter, FC: Fixed carbon,
daf.: Dry ash free, diff. O: Differential O → diff. O = 100 − (C + H + N + S)

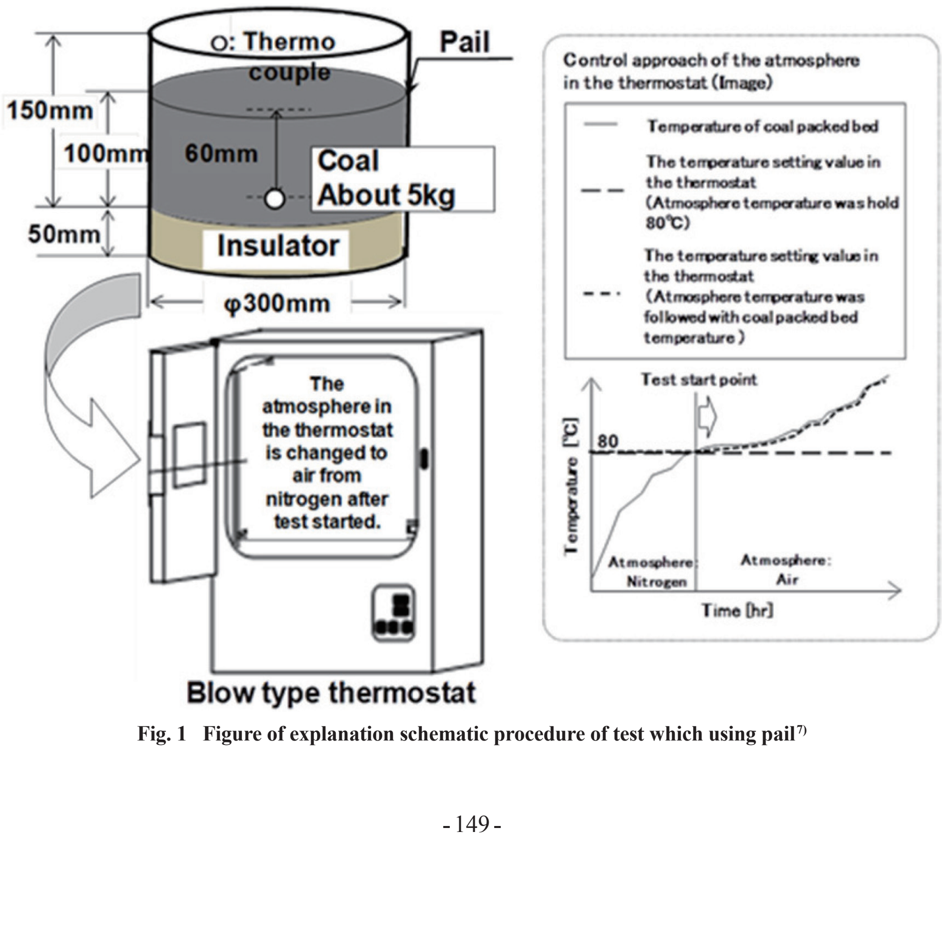

Fig. 1 Figure of explanation schematic procedure of test which using pail[7]

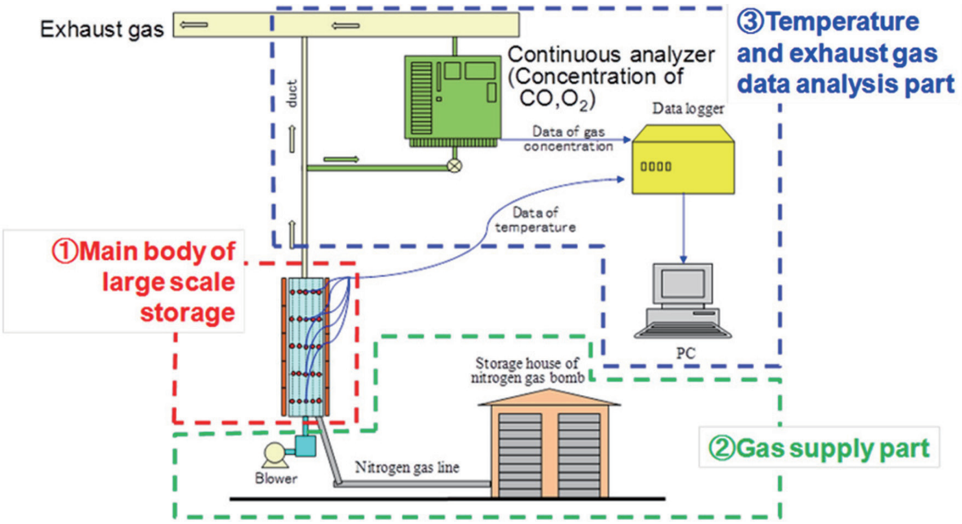

Fig. 2 Configuration conceptual diagram of the evaluation device used for the test[7)]

was used, the tester designed and produced by the Central Research Institute of Electric Power Industry in Japan to verify the storage safety of wood pellets (hereinafter, solid fuel storage safety tester) was employed. The tester was designed simulating a homogeneous storage where heat release to the outside was reduced as much as possible.[9)] **Figure 2**[7)] illustrates the configuration conceptual diagram of the tester. The tester mainly consists of (1) a main tester body with a diameter of 0.5 m and a height of 2 m to store approximately 300 kg of coal samples, (2) gas supply part, and (3) temperature and exhaust gas data analysis part (consisting of an exhaust gas continuous analyzer, device to collect data on the temperature in the coal packed bed (data logger), and PC for recording the data). Nitrogen is supplied from the nitrogen gas supply line and air is supplied from the blower to the bottom of the tester. The gas and air pass through the coal packed bed in the main body and are discharged from the upper section of the tester.

Figure 3[7)] illustrates an outline of the main tester body along with the locations of the thermometers installed inside. Heaters for the gas installed around the supply gas line can warm the gas to be supplied to the preset temperature. Other heaters for samples installed around the main body can control the temperature in the coal packed bed in the tester to the preset temperature. Five heaters for samples have been installed in five levels in the height direction and the temperature of the heater in each level can be separately set. The perimeter of the heaters for samples was covered with an insulator to reduce the influence of the ambient temperature around the tester and such configuration can inhibit the reaction heat generated in the system from being released to the outside air.

As shown in Fig. 3, among the five heaters for samples, each of the second to the fourth levels from the top has five thermometers at the heater height in the middle of the tester in a grid-like fashion. They can measure the temperature in the height direction of the coal packed bed and temperature distribution on the horizontal section over time. The concentration of CO and O_2 in the exhaust gas released from the upper section of the tester was measured with the continuous analyzer (infrared gas analyzer made by Fuji Electric Co., Ltd.).

The test procedures are as follows. The coal samples used were similar to those used for the test using the pail (mean particle size of 3 mm). Approximately 300 kg of the coal samples were charged

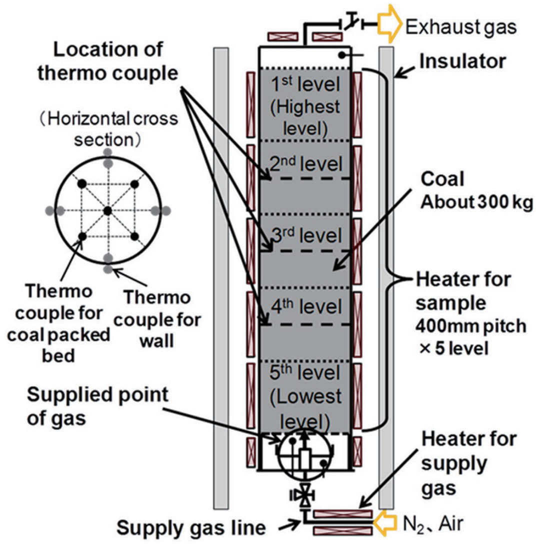

Fig. 3 Schematic diagram of the inside of the device used for the test[7)]

into the tester from the upper section. The coal packing density at that time was approximately 0.8 g/cm^3. Nitrogen gas was supplied from the bottom of the tester (flow rate: 2 L/min) during the temperature increase from room temperature to the test start temperature (60°C) to prevent the coal samples from oxidizing. In the nitrogen atmosphere, the heaters for samples were used for heating until the temperature in the coal packed bed in each level reached 60°C. The test start temperature was determined as 60°C because in the test using the pail, the temperature had not rapidly increased. After confirming that the temperature in the coal packed bed in each level had reached 60°C, the gas supplied from the bottom of the tester was changed from nitrogen to air (flow rate: 2 L/min, hygroscopic moisture: atmospheric humidity). The test was started at that point.

It was considered in this test to closely resemble a state in which coal was stored in a coal bin in actual operation. When coal is discharged from the lower section of an actual coal bin, although air flows in from the outlet, air hardly flows in on other occasions. Therefore, the lower limit of the flow rate value (flow rate: 2 L/min) that this tester could control was determined as the air flow rate. The test was continued for 10 days. To reproduce the insulating conditions, the temperature measured with all the thermometers was monitored during the test and the heater output was adjusted such that the difference between the mean temperature in the coal packed bed and that at the wall of the tester would be within ±1°C, in order to control the temperature in the coal packed bed.

2.2 Results and consideration

2.2.1 SIT test results

Figure 4[7] shows the SIT test results. The figure clearly shows that the sample temperature increase significantly differs depending on the test start temperature. Comparing the results between the test start temperature of 90°C and 100°C, the sample temperature starts increasing at a shorter interval after the test start in the case of the test start temperature of 100°C. Under the test conditions, when the test start temperature was 80°C, no sample temperature increase was seen during the measurement for the three days.

2.2.2 Results of the test using the pail

Figure 5[7] shows the results of the pail test under Condition 1. In the SIT test with the test start temperature of 80°C, no sample temperature increase was observed. In this test, the temperature in the coal packed bed reached 83°C at maximum during the five-day test. After that, it gradually approached 80°C, which was the ambient temperature. This phenomenon may have been caused because oxidation heat generated in the system was released into the atmosphere. Accordingly, to examine the state when the inside of the system was heat-insulated, a test under Condition 2 was performed.

Figure 6[7] shows the test results. A phenomenon where the temperature in the coal packed bed slightly increases and then lowers, as shown in Fig. 5, is not observed. The temperature in the coal packed bed gradually increases from the test start and then it starts rapidly increasing in approximately two days; it reaches 200°C in 2.4 days after the test start. These results show that the inside of a tester that is used to evaluate the heat generation characteristics of oxidized coal needs to be heat-insulated and the results vary depending on the coal sample mass used for the test.

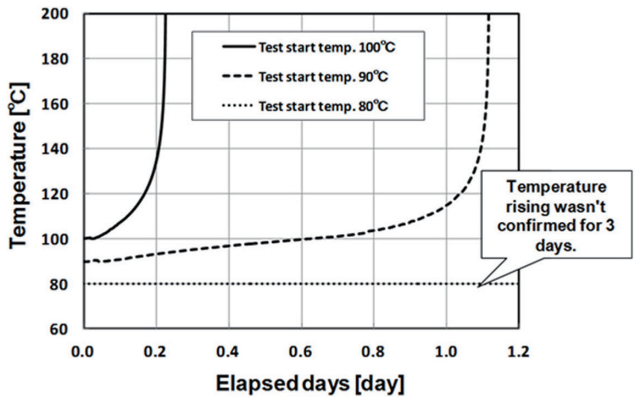

Fig. 4 Result of SIT experiment[7]

2.2.3 Results of the test using the solid fuel storage safety tester

The solid fuel storage safety tester was used to perform a test using coal samples of a few hundred kilograms. **Figure 7**[7] shows the changes in the temperature in the coal packed bed in each level of the tester. In the SIT test using a small quantity of coal samples with the test start temperature of 80°C, the sample temperature remained constant at 80°C (Fig. 4). In this test, even when the test start temperature is 60°C, the temperature in the coal packed bed gradually increases. In addition, no differences are seen in the temperature changes in each of the second to fourth levels in the horizontal direction of the tester and the temperature increases almost evenly. A comparison of the temperature changes in the second to fourth levels shows that the temperature in the coal packed bed in the fourth level obviously increases; as the depth from the surface is deeper, the degree of the temperature increase in the coal packed bed is larger. These results show that in a laminated state like stored coal, the rate of the temperature increase due to heat generation as a result of coal oxidation differs in the height direction of the coal packed bed.

Next, **Fig. 8**[7] shows the comparison results of the temperature changes in the coal packed bed of the fourth level that shows the most obvious temperature increase along with the changes in the exhaust gas components. The O_2 concentration changes in the early stage of the test show that the O_2 concentration gradually approach-

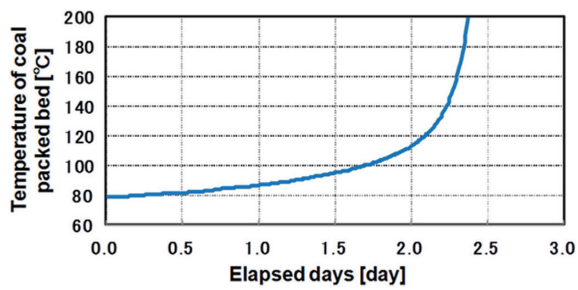

Fig. 6 Test result using a pail with the atmosphere temperature was followed with coal packed bed temperature[7]

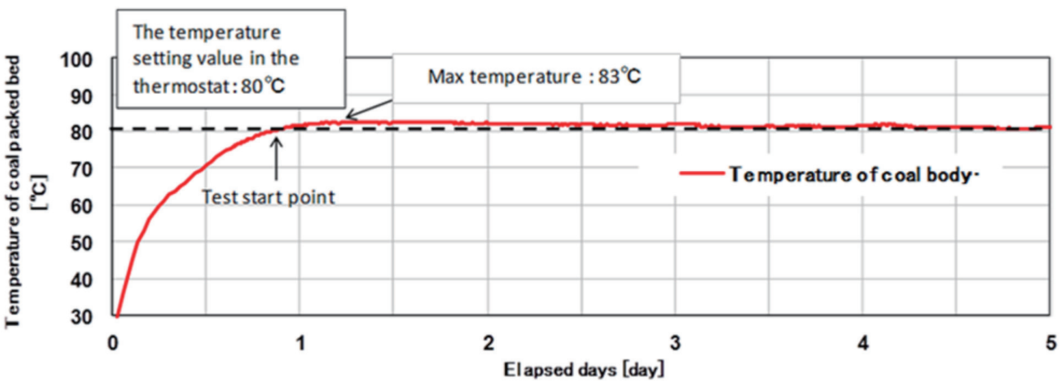

Fig. 5 Test result using a pail with the atmosphere temperature was hold at 80°C[7]

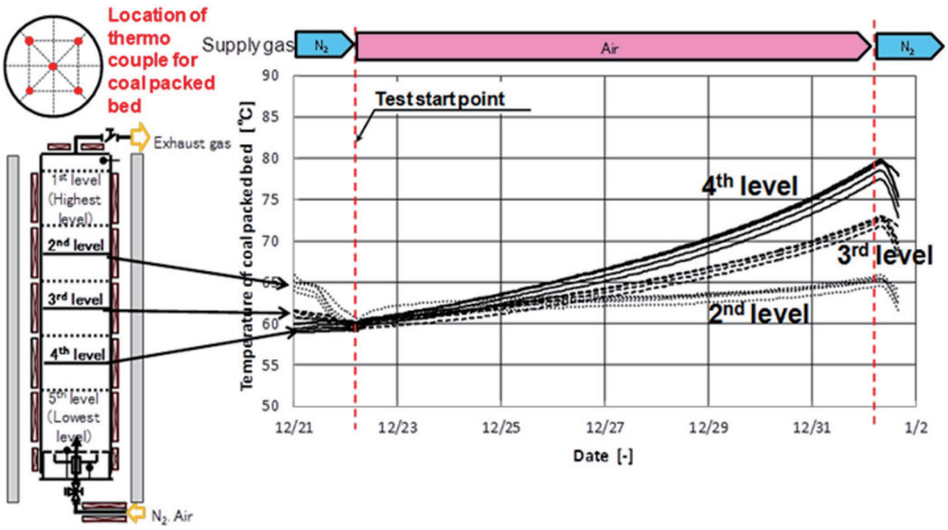

Fig. 7 Transition of temperature at each level of coal packed bed[7)]

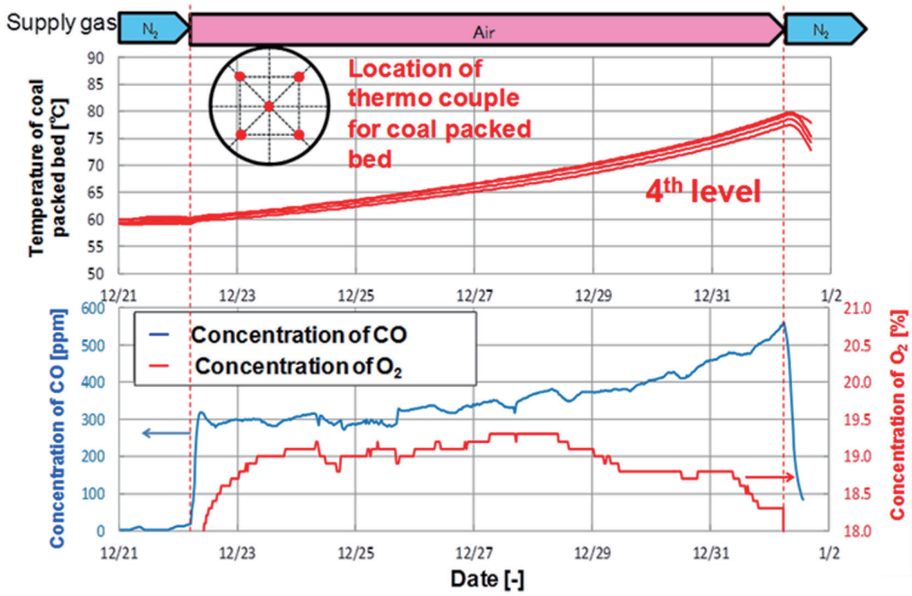

Fig. 8 Transition of temperature at 4th level of coal packed bed and transition of exhaust gas components[7)]

es saturation. The changes in the exhaust gas components show that the CO concentration continuously and gradually increases after the test start, and the O_2 concentration remains constant for approximately six days in the first half of the test period and then it starts decreasing after that. These results show that the temperature increase in the coal packed bed at least in the second half of the test is determined mainly by the reaction heat from the oxidation of the coal.

The SIT test and pail test results above show that the results vary depending on the coal sample mass used for the test and thereby heat insulation inside the system is important. The test using the solid fuel storage safety tester shows that the temperature distribution in the height direction of the coal packed bed needs to be considered based on the target process.

These experimental results show that to evaluate the safety of coal storage, the coal sample mass to be used needs to be considered based on the actual process and the inside of the system needs to be sufficiently heat-insulated to inhibit the heat from being released.

3. Conclusion

In this study, to evaluate the safety of coal storage indoors, various tests were performed with different coal sample mass (a few grams, a few kilograms, and a few hundred kilograms) and different heat insulation of the testers. The findings are shown below.

1) Even under the same temperature conditions, the rate of the temperature increase due to the oxidation of the coal significantly differs depending on the coal sample mass used for the test; even under the condition where the temperature is not increased in the SIT test, it increases in the pail test.
2) Even with the same coal sample mass, the temperature increase due to the oxidation of coal accelerates and proceeds under the condition where the heat insulation state in the system is maintained as compared with the condition where the temperature of the surrounding area is maintained as constant at the initial

setting temperature.

3) Even when the adiabatic state is maintained inside the system, the temperature increase characteristics vary depending on the depth in the coal bed.

This study evaluated the heat generation characteristics of the coal in the three methods. The results show that each method has both advantages and disadvantages. The test using the small sample mass produced the results in a simple and quick way while the heat generation characteristics tended to be underestimated. The tests using the large sample mass could evaluate the heat generation characteristics precisely while they took labor and time to obtain the results.

At coke production sites, a few types of coal grades are mixed and the quality often varies for each time of delivery even in the same grade. To use such coal safely, the oxidation and heat generation characteristics of each coal grade need to be accurately and quickly evaluated. To that end, the advantages and disadvantages of each evaluation method may need to be considered and combined for evaluation.

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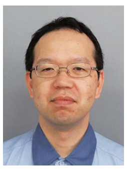

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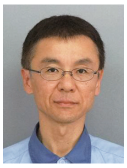

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