Carbon Deposition Mechanism in Coke Oven Chamber – Influence of Fine Particles on Formation Rate of Carbon Deposits –

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Abstract

Particles originated from coal are included in carbon deposits collected from a coke oven chamber. The amounts of such particles are much greater in dry coal operation than in wet coal operation. In the present study, the experimental study was carried out to evaluate the influence of coal fines on the pyrolysis reaction of the coal carbonization gas. The reaction rate increases when coal fines coexist with coal carbonization gas at the temperature from 950 to 1250K. Dust generation test was also performed in the study. It was found that dust generation rate of coal charge changes depending on the coal moisture, size distribution, graindability, and so on. Furthermore, the equation for estimating pyrolysis reaction rate of carbonization gas in considering coal fines was derived based on these experimental data.

1. Introduction

When coal is heated (carbonized) in a coke oven, hydrocarbon-based gas (coal carbonization gas) is generated. The coal carbonization gas passes through the upper space of the coke oven chamber (oven top space) and is exhausted from the oven and sent to the succeeding process. Since the oven top space is a high temperature atmosphere (1,100 to 1,200K), the coal carbonization gas is subject to thermal decomposition there, causing carbon to deposit on the surface of the refractory bricks that make up the coke oven chamber. If the amount of carbon deposits becomes excessive, it not only reduces the effective volume of the coke oven chamber, thereby reducing productivity, but also increases the resistance of the coke when it is pushed out of the chamber. Thus, excessive carbon deposits can be an impediment to stable operation of the coke oven battery.

Photo 1 is a sectional view of carbon deposits collected from a coke oven chamber, observed under a polarizing microscope. In addition to carbon as a product of the thermal decomposition of coal carbonization gas, inert components of the coal can be observed. It
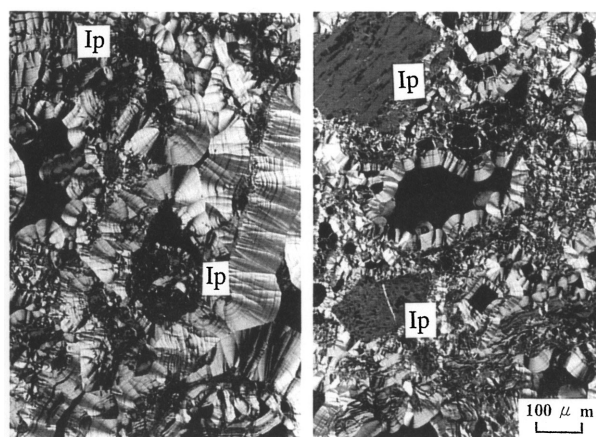
Photo 1 Polarized light micrograph of carbon deposit collected from coke oven chamber (Ip: inert component from coal)

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is considered that those inert components are dust particles that were generated during the charging of coal into the coke oven chamber and embedded in the carbon deposit during the carbon formation process by the thermal decomposition of coal carbonization gas. According to Furusawa et al.[1], carbon deposits in the oven top space from dry coal operation (moisture content of charging coal: 6 to 7%) contains 10 to 20% of coal-originated components, more than twice the proportion from wet coal operation (moisture content of charging coal: 9 to 10%).

The carbon formation process by the thermal decomposition of coal has been studied by Jomoto et al.[2], Nagata et al.[3] and Dumay et al.[4]. However, there are few reports on studies of the formation of carbon deposits with consideration given to the influence of dust particles originating from the coal[1,5]. A study was conducted on the influence coal-originated dust particles have on the formation of carbon deposits from coal carbonization gas in the top space of a coke oven chamber. Then, a formula for estimating the rate of carbon deposit formation was devised. This paper describes the results of the present study.

2. Experimental Apparatus and Procedure
2.1 Experiment to evaluate influence of dust particles
The experimental apparatus used is presented schematically in **Fig. 1**. A carbonization furnace (material: SUS 304) measuring 140 mm (W) × 500 mm (L) × 600 mm (H) was heated to approximately 900℃ and coal was fed into the furnace via a charging hole at the furnace top. Coal carbonization gas generated in the furnace was directed into a pyrolysis reactor through a horizontal tube (50 mm in diameter) which was kept hot by a heater. In the reactor, the coal carbonization gas was brought into contact with a piece of silica brick (dimensions: 6 × 20 × 60 mm) to form carbon deposit.

With a vibrating-type fine coal feeder installed above the silica brick, coal whose particle size was pre-adjusted was fed in at a prescribed rate. The amount of carbon generated by the thermal decomposition of coal carbonization gas was calculated from the change in weight of the silica brick.

Charging coal (5.6% moisture, 27.0% volatile matter) as used in an actual coke oven was adopted as the coal for carbonization and the fine coal. The charging coal was crushed so that the proportion of particles 3 mm or less in size (−3 mm) was 78% for the coal for
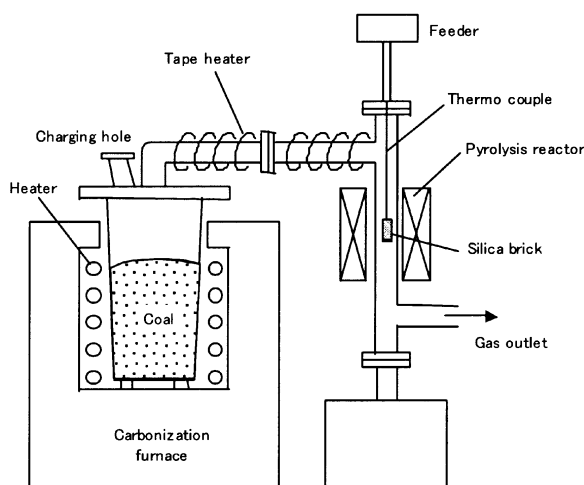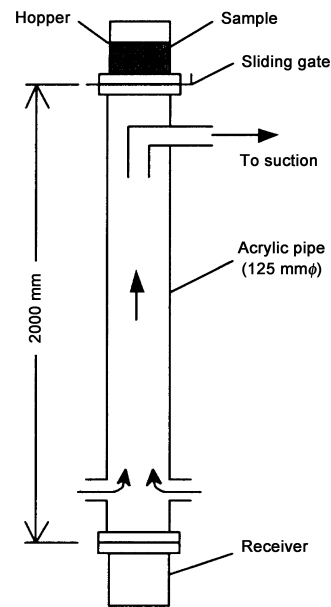

Fig. 2 Experimental apparatus for dust test

carbonization, while the proportion of particles 0.1 mm or less in size (−0.1 mm) was 98% for the fine coal. The coal carbonization time was 180 minutes.
2.2 Experiment to Evaluate Dusting Characteristics
In this experiment, an acrylic cylinder (125 mm in ID, 2,000 mm in height) as shown in **Fig. 2** was used. With a suction tube inside the acrylic cylinder connected to an outside decompression line, an ascending air current (1 meter/sec) is continually passed through the tube. The sample (1 kg of coal) in the hopper at the top of the cylinder is fed into the cylinder through the sliding gate and sucked by the suction tube for one minute. The dust ratio, Rd, was defined as shown in Equation 1.

$$Rd\ (\%) = 100\ [(\text{amount of sample put in} \\ -\ \text{amount of sample recovered by receiver}) / \\ \text{amount of sample put in}] \qquad (1)$$

As samples, coal brands whose Hardgrove Grindability Index (HGI)[6] is in the range 53 to 90 were selected and adjusted to −3 mm particle size 70 to 90%, moisture 0 to 3% and oil addition 0 to 3%.

3. Experimental Results and Discussion
3.1 Influence of fine coal on carbon deposition rate
3.1.1 Influence of temperature on deposition rate
The relationship between carbon deposition rate and temperature is shown in **Fig. 3**. It can be seen that when both coal carbonization gas and fine coal exist (curve indicated by ◯), the carbon deposition rate is much higher than when there is only coal carbonization gas (curve indicated by △). It can also be seen that the carbon deposition rate is lowest at a temperature of around 1,000K when both coal carbonization gas and fine coal are present (around 900K if only coal carbonization gas): it is higher at other temperatures. The increase in carbon deposition rate on the low-temperature side is considered to be due to the condensation of tar contained in coal carbonization gas on the surface of the brick, rather than the thermal decomposition of coal carbonization gas.
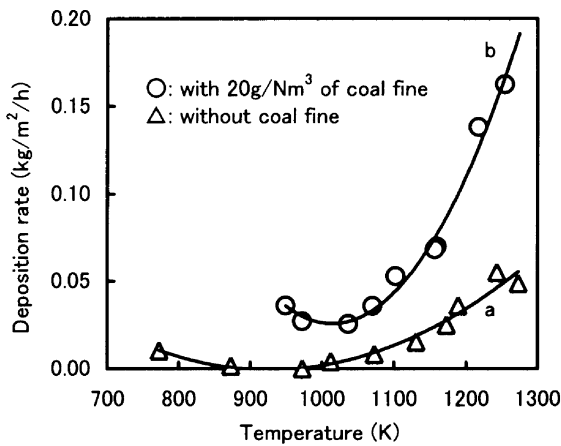

Fig. 1 Experimental apparatus for carbon deposition

Fig. 3 Influence of temperature and fine on carbon deposition rate

Fig. 5 Comparison of experimental data with actual one

3.1.2 Influence of fine coal concentration on carbon deposition rate

The relationship between fine coal concentration and carbon deposition rate at 1,173K is shown in **Fig. 4**. It can be seen that the higher the fine coal concentration, the higher the carbon deposition rate. It should be noted, however, that the relationship between them is not linear: the carbon deposition rate increases at a faster pace than does the fine coal concentration. The reason for this is probably that the higher the fine coal concentration, the larger the number of irregularities on the brick surface and the greater the surface area that takes part in the deposition of carbon.

3.1.3 Compatibility of experimental results with actual coke oven measurements

Fig. 5 shows the results of a comparison between the experimental results shown in Fig. 3 and the carbon deposition rates measured in the top space of a coke oven in dry coal operation (charging coal moisture: 5.7%). In this figure, the solid lines, a and b, correspond to the approximate curves, a and b, in Fig. 3 representing the experimental results. The black circles indicate measured values obtained in an actual coke oven, and each of the figures in parentheses indicates a measuring time (elapsed time from completion of charging
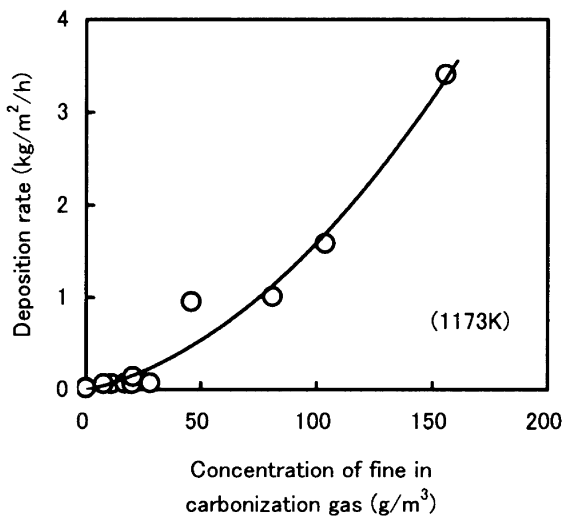
of coal into coke oven chamber). The fine coal concentration in the coke oven chamber was 20 g/Nm^3 one hour after charging of coal and 5 g/Nm^3 two hours after charging of coal. Thus, the fine coal concentration sharply decreases after the charging of coal[1]. It is considered that six hours and more after the charging, the deposition of carbon is due mainly to the thermal decomposition of coal carbonization gas. Therefore, the experimental results can be judged valid from the knowledge that has been obtained from an actual coke oven.

3.2 Dusting characteristic of coal at time of charging

It was found that coal dust occurs when coal is charged into a coke oven chamber and that such coal dust significantly influences the rate of carbon deposition from coal carbonization gas. Therefore, a laboratory experiment was conducted to quantitatively grasp the behavior of coal dust generation. Namely, using four types of coals having different HGI values, the dust ratio, Rd, of each was measured while varying the moisture, particle size and oil addition. The experimental results are shown in **Table 1**. Assuming that the Rd could be expressed as a linear combination of HGI, coal particle size (-3 mm%), coal moisture (Mois) and oil addition (Oil%), calculations were made as coefficients for each of the terms by the method of least squares. As a result, the following equation was obtained.

$$Rd\ (\%) = 0.02\ [HGI] + 0.02\ [-3mm\%] - 0.26\ [Mois]$$
$$- 0.22\ [Oil\%] - 0.99\ (R^2 = 0.57) \qquad (2)$$

HGI: Hardgrove grindability index
-3 mm%: Weight percentage of coal particles 3 mm or less in size (mass %)
Mois: Coal moisture (mass %)
Oil%: Oil addition to coal (mass %)

3.3 Establishment of formula for estimating carbon deposition rate

On the basis of the above experimental results, a formula was created for estimating the rate of carbon deposition that reflects the influence of fine coal on the carbon deposition rate.

3.3.1 Equation that takes fine coal concentration into account

The experimental results shown in Fig. 3 were Arrhenius-plotted as shown in **Fig. 6**. The activation energy calculated from the inclination of the straight line is 10.6 kJ/mol when both coal carboniza-

Fig. 4 Influence of concentration of fine on carbon deposition rate

Table 1 Results of dust test

		Coal A (HGI: 89.6)							Coal B (HGI: 53.4)						
Moisture	(%)	0			3			0	3	0			3		0
Oil addition	(%)	0			0			3	3	0			0		3
Under 3mm	(%)	71.8	79.6	88.3	69.6	79.0	87.8	78.5	77.6	71.8	81.3	89.4	70.2	78.1	88.6	77.8	80.1
Dust ratio	(%)	2.33	2.93	2.90	0.40	0.74	0.79	1.08	1.11	1.09	1.27	1.31	1.02	0.96	1.67	0.45	0.60

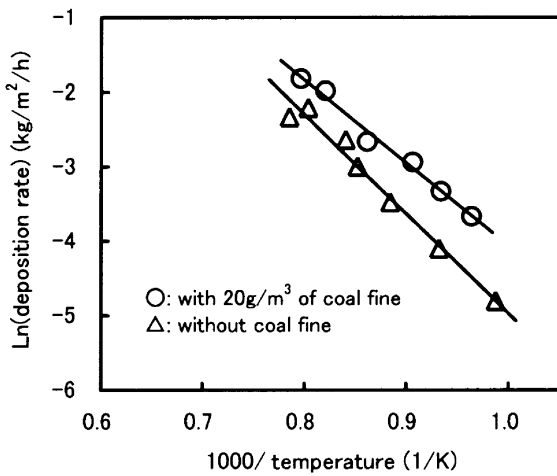|		Coal C (HGI: 85.5)				Coal D (HGI: 54.2)	
Moisture	(%)	0		3		0	3
Oil addition	(%)	0		0		0	0
Under 3mm	(%)	74.1		72.8	76.2	86.3	81.2	74.4
Dust ratio	(%)	2.43		1.21	1.27	1.56	1.36	0.55

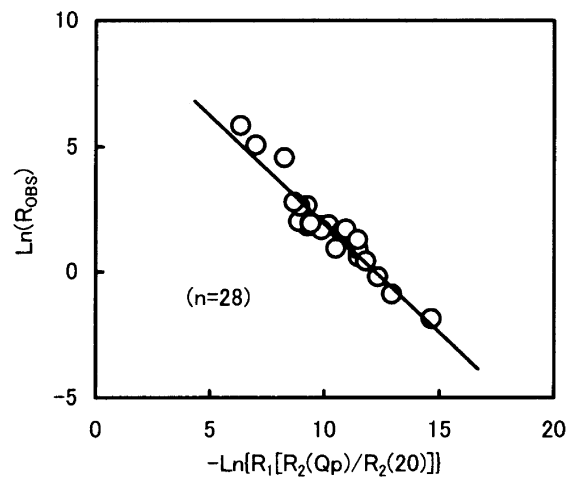**Fig. 6 Arrhenius plots for carbon deposition rate**

Fig. 7 Relationship between experimental data and calculation results by Equation (5)

tion gas and fine coal are present and 11.2 kJ/mol if there is only coal carbonization gas. Since the two activation energies are nearly the same, their average value was used to express the influence of temperature on the rate of carbon deposition as follows.

$$R_1 = k_1 \exp(-10,900 / T) \qquad (3)$$

R_1: Carbon deposition rate (kg/m^2/h)
k_1: Coefficient
T: Temperature (K)

By approximating the relationship between the carbon deposition rate and fine coal concentration shown in Fig. 3 as a quadratic expression, the following equation can be obtained.

$$R_2 (Qp) = 0.01Qp^2 + 0.593Qp + 1.98 \qquad (4)$$

R_2: Carbon deposition rate (kg/m^2/h)
Qp: Fine coal concentration in coal carbonization gas (g/m^3)

From Equation 3 representing the influence of temperature and Equation 4 representing the influence of fine coal, the following equation was derived that takes into account the influences of temperature and fine coal concentration. It should be noted that the influence

of fine coal concentration incorporated in the equation is the influence observed when Qp = 20 g/Nm^3 in the present experiment.

$$R_3 = k_2 R_1 [R_2(Qp) / R_2(20)] \qquad (5)$$

R_3: Carbon deposition rate (kg/m^2/h)
k_2: Coefficient

There is a good linear relationship between the calculated values obtained by Equation 5 and the experimental results as shown in **Fig. 7**. Therefore, Equation 5 can be rewritten as follows.

$$Ln (R_{OBS}) = 0.86Ln [R_1 * R_2(Qp) / R_2(20)] + 10.6 \qquad (6)$$

R_{OBS}: Carbon deposition rate obtained experimentally (kg/m^2/h)

3.3.2 Equation that takes coal properties into account
Concerning the influence of moisture and volatile matter in coal on the rate of carbon deposition from coal carbonization gas, the following equation has been reported by Jomoto et al[2].

$$Do = 64.5\exp(-7,950 / T) VM (1 - 0.0476 Mois) \qquad (7)$$

Do: Carbon deposition rate (mm/day)
VM: Volatile matter in coal (mass% - dry)

T: Temperature (K)

Mois: Moisture content of coal (mass%)

Here, the coefficient C_f was considered that takes into account the influence of any change in coal properties at a given temperature. Using the carbon deposition rate for the coal properties (27% volatile matter, 5.6% moisture) in the present experiment as the reference and calculating carbon deposition rates for different coal properties as ratios to the reference, C_f can be expressed as follows.

$$C_f = 0.051 VM (1 - 0.0476 \, Mois) \quad (8)$$

By multiplying Equation 6 that takes into account the influence of fine coal by coefficient C_f obtained by Equation 8, we obtained the following equation that takes into account the influences of coal properties and temperature.

$$\begin{aligned} R_{EST} &= C_f * R_{OBS} \\ &= k_3 VM(1 - 0.0476 Mois) * [0.01Qp^2 + 0.593Qp + 1.98]^{0.86} \\ &\quad * [exp \, (- 10{,}900 \, / \, T)^{0.86} \quad (9) \end{aligned}$$

R_{EST}: Estimated carbon deposition rate (kg/m³/h)

k_3: Coefficient

Using the value of Rd obtained by the dust rate measurement, fine coal concentration Qp can be calculated by the following equation.

$$Qp \, (g/Nm^3) = [Rd \, / \, 100] \, / \, [3.14 Ra^2 V_f t_s] \quad (10)$$

Ra: Cylinder radius (m)

V_f: Ascending air current velocity in suction tube (m/s)

t_s: Time for which fine coal is generated (s)

By using Equations 2, 9 and 10, it is possible to estimate the rate of carbon deposition taking into account the influence of fine coal that occurs when coal is charged.

3.4 Study of influences of various factors using estimation equation

Figs. 8 (a) through (d) show examples of the influence of various factors on the rate of carbon deposition in the oven top space that were estimated using Equation 9. It should be noted that although in an actual coke oven the temperature and fine coal concentration vary with time, it was assumed, for the purpose of simplifying the calculations, that the temperature and fine coal concentration (average values) remained steady.

Fig. 8 (a) shows the influence of temperature on the carbon deposition rate. It can be seen that the carbon deposition rate increases with the rise in temperature and that the lower the moisture content of coal, the greater the influence of temperature. The implication is that temperature control is extremely important in dry coal operation.

As shown in Fig. 8 (b), the carbon deposition rate increases as the moisture content of charging coal decreases, and the influence of moisture content is greater at higher temperatures. The figure also shows the influence of oil addition. It is estimated that the carbon deposition rate can be lowered by oil addition to restrain the dust ratio of coal from increasing when the moisture content decreases. It should be noted, however, that since some types of oil promote carbon deposition as does the coal carbonization gas[7], it is considered undesirable to add too much oil.

Fig. 8 (c) shows the influence of the coal particle size. The larger the proportion of coal particles of 3 mm or less (− 3 mm%), or the smaller the coal particle size, the higher the carbon deposition rate
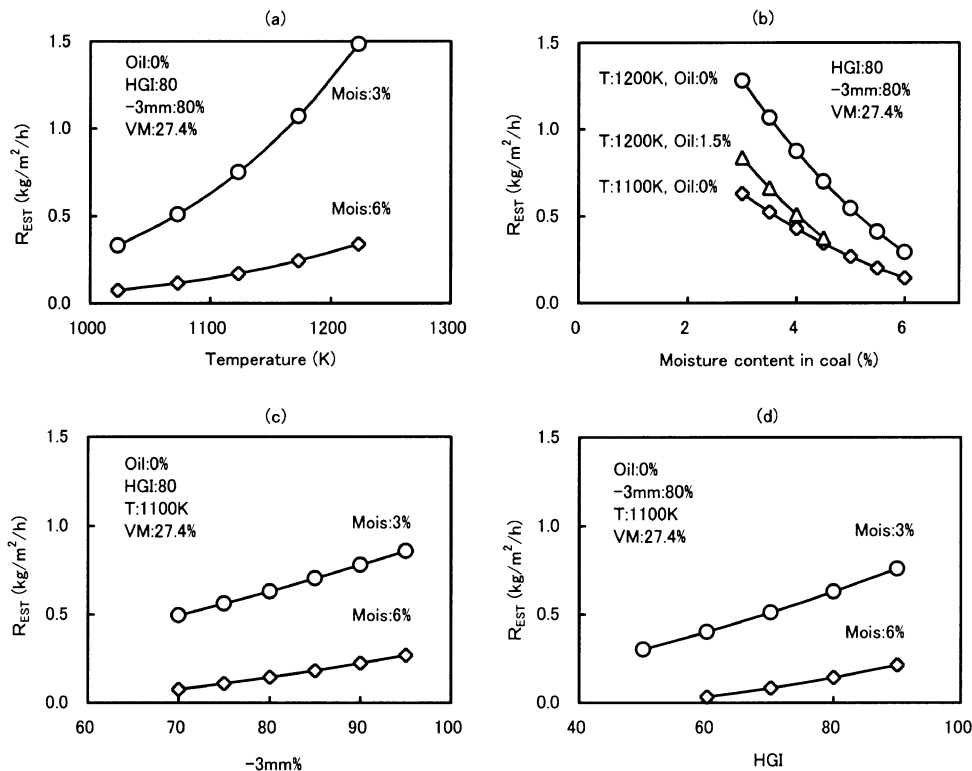

Fig. 8 Evaluations of carbon formation rate by equation (9)

because the dust ratio of coal increases. This is also true of the influence of HGI shown in Fig. 8 (d). It is estimated that when brittle coal (coal of large HGI) is used in large quantities, carbonaceous matter that enters the carbon deposit from the fine coal will increase.

4. Conclusion

An experimental study was made of the influence of fine coal (coal dust) that occurs during the charging of coal on the carbon deposition rate, which significantly affects the stable operation of a coke oven battery. As a result, it was confirmed that when both coal carbonization gas and fine coal are present, the carbon deposition rate increases in quadratic function manner relative to the fine coal concentration. In addition, a laboratory experiment was conducted to evaluate the dusting characteristics of various charging coals and formulated them in terms of HGI, coal particle size, moisture content and oil addition rate. Furthermore, by combining the results of the present experiment and the conventional equation on the rate of carbon deposition from coal carbonization gas, a formula was produced for estimating the rate of carbon deposition when both coal carbonization gas and fine coal are present.

References
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