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| Title            | A Theory on the Mechanism of an Outbreak of Spontaneous Combustion of Coal II   |
| Author(s)        | Hashimoto, Kiyoshi  |
| Citation         | Memoirs of the Faculty of Engineering, Hokkaido University, 11(3), 269-291      |
| Issue Date       | 1962-03   |
| Doc URL          | <a href="http://hdl.handle.net/2115/37824">http://hdl.handle.net/2115/37824</a> |
| Type             | bulletin (article)  |
| File Information | 11(3)_269-292.pdf   |



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# A Theory on the Mechanism of an Outbreak of Spontaneous Combustion of Coal II

Kiyoshi HASHIMOTO

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## I. Preface

In this paper the theory on the mechanism of an occurrence of spontaneous combustion of coal which was presented in the preceding paper by the same author<sup>1)</sup> is developed. Changes in the heat generation caused by oxidation according to the quantity of coal—the residual coal in a mine or storage coal—are discussed hereinafter.

The quantity of coal chiefly influences the preservation and conduction of heat and the control of oxidation. In order to investigate these phenomena, however, the amount of air leakage must be known. As far as the present writer is aware, it is at present impossible to measure this in the underground.

In “Experimental Studies of the Leakage through Wastes” (Japanese) published formerly by the present writer<sup>2)</sup>, experiments under conditions in which spontaneous combustion is most possible were conducted in order to measure the approximate quantity of air leakage caused by pressure differences. This kind of air leakage is one of the most serious factors in spontaneous combustion in the underground. Besides pressure differences, leakage of air caused by tempera-

ture differences and the production of methane gas, directly cause spontaneous combustion both in the underground and elsewhere.

In Furnas's report<sup>3)</sup>, natural air circulation caused by differences of temperature has already been discussed, and by the same writer<sup>4)</sup> leakage of air caused by generation of methane gas has been considered.

The heat generation phenomena due to oxidation of coal stored on the surface are chiefly caused by natural ventilation, but at the early stages of oxidation, they are caused by the production of methane gas alone. Consequently in this report Furnas' final equation is quoted, together with a partial substitution concerning methane gas.

In this paper the heights of stored broken coal are assumed to be two, four and six meters. These figures are significant in deciding the heights of bunker, if the effects of downward cooling in the environment of the coal are taken into consideration.

In the case of particle size coal the phenomenon of heat generation by oxidation shows that there is little air circulation, while the heat generation in big grain coal shows that there is much air circulation.

## II. Flow of air through beds of solid particles of coal

### (1) Circulation of air and gases caused by temperature difference

Warm air rises. Air will rise if its density is less than that of the surrounding atmosphere. The cause of this movement is the difference of pressure.

According to Mingle the weight of one centimeter of air column at an optional temperature is

$$\frac{1.265}{1000} \times \frac{273}{T} = \frac{0.345}{T}$$

Therefore the difference of weight between the two optional absolute temperatures of  $T_1$  and  $T_2$  is

$$0.345 \left[ \frac{1}{T_1} - \frac{1}{T_2} \right]$$

That is to say, the difference of pressure which causes the movement of air is

$$\Delta P_{\infty} = 0.345 \left[ \frac{1}{T_1} - \frac{1}{T_2} \right] \quad (1)$$

cm. of water column per cm. of air column.

$T_1$ : temperature of air

$T_2$ : temperature of coal

When  $T_1$  is 298°K (25°C), and  $T_2$  is 273+50°C, 273+100°C, 273+150°C and 273+200°C, and if the atmospheric pressure is 760 mmHg,  $\Delta P_\infty$  is as follows:

|              |                              |
|--------------|------------------------------|
| 25°C ~ 50°C  | $\Delta P_\infty = 0.000089$ |
| 25°C ~ 100°C | $\Delta P_\infty = 0.000233$ |
| 25°C ~ 150°C | $\Delta P_\infty = 0.000342$ |
| 25°C ~ 200°C | $\Delta P_\infty = 0.000428$ |

If Furnas' experiments<sup>35)</sup> of 1925 concerning a packed tower are applied here, the following experimental equation about the movement of air develops.

$$\Delta P_\infty = \frac{82.8 \times 10^a}{P_d^{1.68} - V_n} \left( \frac{T}{298} \right)^b R^{B(\text{air}, t^\circ)} \quad (2)$$

Where  $a = B_{(\text{air}, 25^\circ)} + 3.9093 - 3.4414 \frac{F}{V_n} + 4.2 V + 0.6177 \left( \frac{F}{V_n} \right)^2 - 8.9 V^2$ ,

$$b = 1.284 - 0.568 B_{(\text{air}, 25^\circ)}$$

$$B_{(\text{air}, 25^\circ)} = \frac{0.006 + d}{0.036 + 0.51d}$$

$$B_{(\text{air}, t^\circ)} = 2.185 B_{(\text{air}, 25^\circ)} - 0.593 - 0.479 [B_{(\text{air}, 25^\circ)} - 0.5] \times \log_{10} T$$

$\Delta P_\infty$ : in centimeters of water per centimeter in a column

$T$ : in degrees Kelvin,  $t$  in degrees centigrade

$t$ : °C

$P$ : total pressure on the system in centimeters of a mercury column (for air at sea level,  $P=76.0$ )

$R$ : the flow rate in liters per second (N.T.P.) per square centimeter of cross sectional area

$V_n$ : the normal voids, expressed as a decimal fraction

$V$ : the actual voids, also as a decimal fraction

$d$ : the equivalent spherical diameter of particles, in centimeters

$F$ : shape factor

This equation applies to dry particles, ranging from 0.01 to 20 cm. in diameter and between 25° and 660°C in temperature. It will be noted that Furnas' equation is of the form,

$$\Delta P_\infty = AR_s^B$$

$\Delta P_\infty$  can be calculated at various temperatures. Therefore when the conditions are changed and the constants  $A$  and  $B$  are known,  $R$  can be determined.

When the diameters of ball-shaped coal are 2 cm., 1 cm., 0.5 cm., 0.25 cm., 0.2 cm. and 0.1 cm., the numerical values of  $\Delta P_\infty = AR_s^B$  are as follows:

When the diameter is 2 cm.,

|       |  |
|-------|--|
| 25°C  | $\Delta P_{\infty} = 13.79 R_s^{1.9}$    |
| 50°C  | $\Delta P_{\infty} = 14.12 R_s^{1.875}$  |
| 100°C | $\Delta P_{\infty} = 14.44 R_s^{1.838}$  |
| 150°C | $\Delta P_{\infty} = 14.82 R_s^{1.796}$  |
| 200°C | $\Delta P_{\infty} = 15.16 R_s^{1.763}$  |
| 25°C  | $R_s = 0$ l/day·cm <sup>2</sup> (N.T.P.) |
| 50°C  | $R_s = 147$ "                            |
| 100°C | $R_s = 210$ "                            |
| 150°C | $R_s = 226$ "                            |
| 200°C | $R_s = 226$ "                            |

In the same way one can get the following Table 1 and Figure 1.

TABLE 1. Circulation of air and gases caused by the difference of temperature

| Diameter of ball-shaped coal<br>cm |                             | Ascending air current l/day·cm <sup>2</sup> (N.T.P.) |      |      |      |      |      |
|------------------------------------|-----------------------------|--|------|------|------|------|------|
|                                    |                             | 2  | 1    | 0.5  | 0.25 | 0.2  | 0.1  |
| Temperature<br>of air 25°C         | Temperature<br>of coal 25°C | 0  | 0    | 0    | 0    | 0    | 0    |
|                                    | " 50°C                      | 147  | 76.8 | 37.1 | 11.5 | 6.9  | 0.58 |
|                                    | " 100°C                     | 210  | 110  | 55.6 | 16.2 | 10.6 | 0.94 |
|                                    | " 150°C                     | 226  | 118  | 57.4 | 18.4 | 11.3 | 1.07 |
|                                    | " 200°C                     | 226  | 116  | 56.7 | 17.9 | 11.1 | 1.00 |

## (2) Circulation of air caused by methane gas in the air

When there is methane gas in the air, the density of the air is low and it ascends; this is called self-ventilation.

The air ascends vertically in the same way as ascending air caused by the rise of temperature does, but the ascending direction of air and gases is not necessarily the same as that of air leakage caused by differences in atmospheric pressure in the underground. In Hokkaido coal mines which have many inclined coal seams, and a lot of gas production, the phenomenon of ascending air caused by methane gas cannot be disregarded.

Figure 2 shows the specific gravity of the mixed gas and the methane concentration, the temperatures indirectly indicating the quantity of the air and gas in the ascending current when the particle diameter of coal is 0.5 cm. For example, when the temperature is 25°C and the methane concentration is 40%, the specific gravity is 0.755 (N.T.P.), the equivalent temperature to air quantity is 89°C and the ascending current is 53 l/day·cm<sup>2</sup>. Therefore the volume of intake air is 32 l/day·cm<sup>2</sup> and that of the gas is 21 l/day·cm<sup>2</sup>.

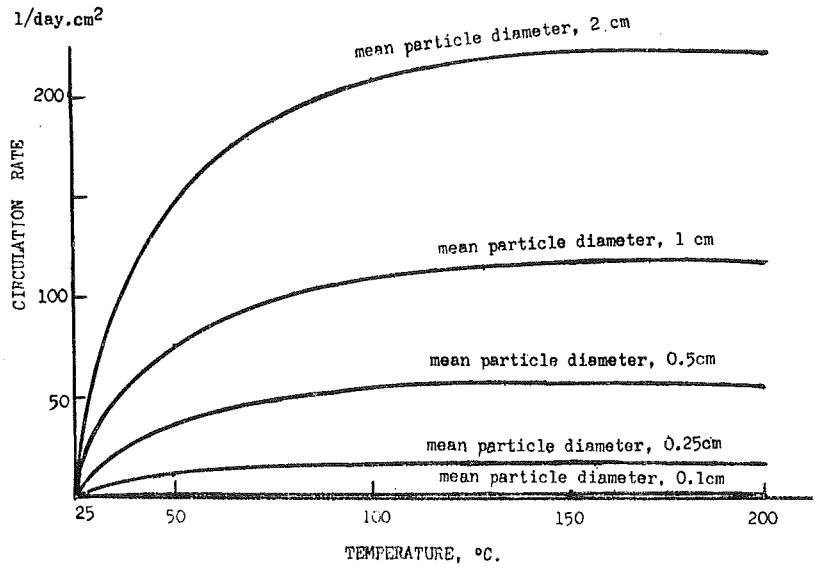


Fig. 1. Circulation of air and gases caused by the difference of temperature

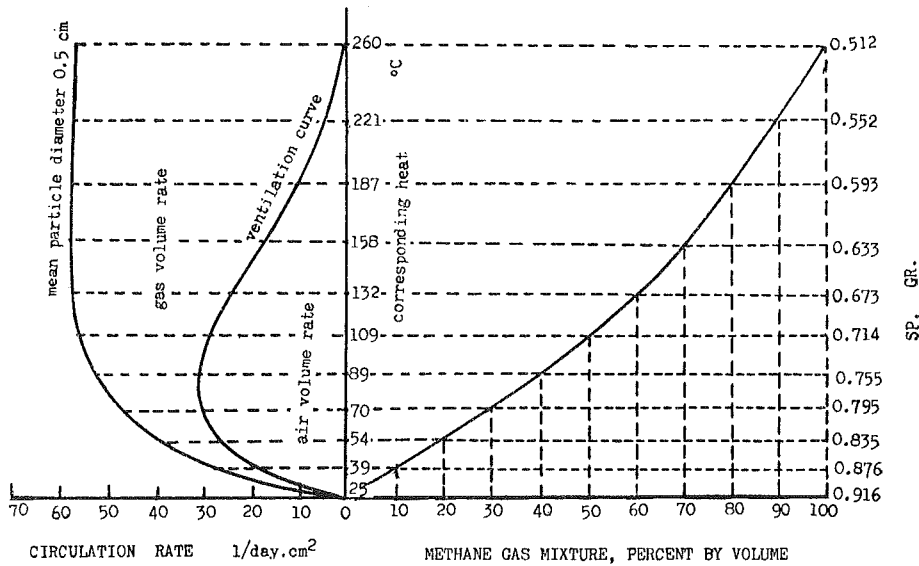


Fig. 2. Relation between percent of methane gas and circulation rate  
average particle diameter 0.5 cm  
temperature of atmosphere 25°C

In any other case with the exception of material with a 0.5 cm diameter one can work out a similar graph in the same way. According to Figure 2

the volume of intake air is maximum when the density of the gas is 30~40%. This seems to explain the fact that the spontaneous combustion of coal could not be prevented at the Akabira coal mines in Hokkaido when the gas concentration in the gob was under 30%.

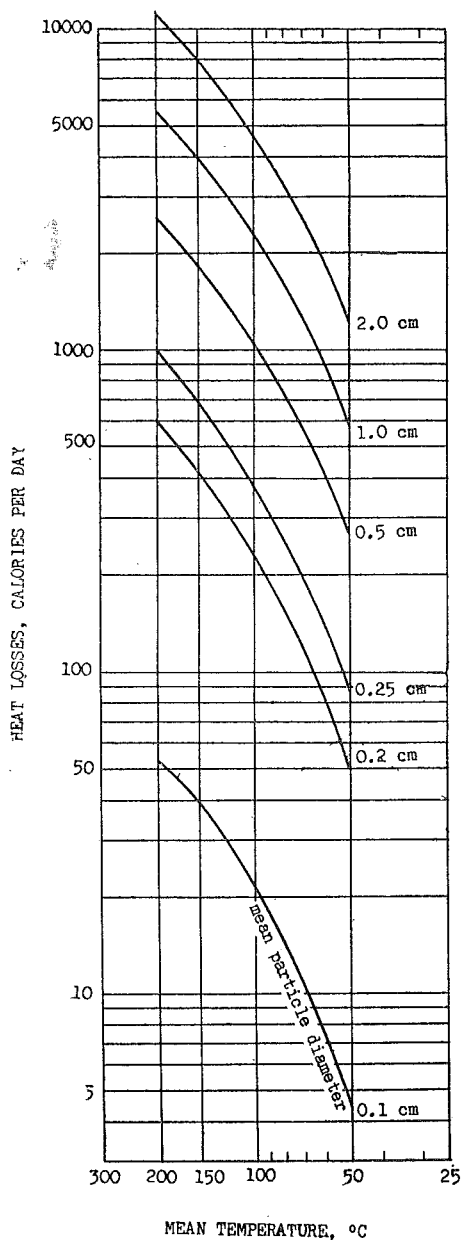


Fig. 3. Heat loss in various sizes of coal

### III. Dissipation of heat

When oxygen is supplied into coal, it is always ventilated with air. When there is ventilation, oxidated gas is always formed. Highly heated coal always loses heat, while high temperature gas or air gives heat to low temperature coal.

In the preceding paper the existence of heat generation phenomena due to oxidation within the area between the perfect heat accumulation curve and the perfect heat radiation curve was noted. In this paper to determine the rate of temperature rise, heat dissipation was quantitatively and minutely measured.

Heat loss to circulating air and gas is calculated according to the following formula of Scott when the specific heat at a given air pressure is  $C_p = 0.24$  cal/°C (0°C~100°C) and the mean specific heat of air per mol is  $C_{pm} = 6.9$  cal.

$$\left(\frac{dQ}{d\theta}\right)_L = \frac{R}{22.4} \times C_{pm} \times \Delta t \quad (3)$$

where

$$\left(\frac{dQ}{d\theta}\right)_L : \text{heat loss, in cal/day} \cdot \text{cm}^2$$

$R$  : the flow rate in  $\text{l/day}\cdot\text{cm}^2$

$C_{pm}$ : means specific heat of air per mol between  $25^\circ\text{C}$  and the temperature of the exit gases

$\Delta t$  : difference in temperature between the air entering the pile and the gases leaving it.

When the temperature of the entering air is  $25^\circ\text{C}$  and the particle diameter of the coal is 2.0 cm, heat loss according to the temperature of inflow air is as follows:

| Temperature of coal | Heat loss                                |                           |
|---------------------|--|---------------------------|
|                     | cal/day $\cdot\text{cm}^2$               | cal/hr $\cdot\text{cm}^2$ |
| $25^\circ\text{C}$  | 0  | 0                         |
| $50^\circ\text{C}$  | $\frac{147}{22.4} \times 6.9 \times 25$  | 1130                      |
| $100^\circ\text{C}$ | $\frac{210}{22.4} \times 6.9 \times 75$  | 4850                      |
| $150^\circ\text{C}$ | $\frac{226}{22.4} \times 6.9 \times 125$ | 8700                      |
| $200^\circ\text{C}$ | $\frac{226}{22.4} \times 6.9 \times 175$ | 12180                     |

Heat loss in various sizes of coal is shown in Figure 3.

#### IV. Effects of concentration of oxygen on oxidation rate

There have been many studies on the effects of the concentration of oxygen on the oxidation rate in the process of oxidation in coal. Winmill<sup>9)</sup> stated that the rate of oxidation of pyrite is directly proportional to the concentration of oxygen. For English bituminous coals, however, he states that the rate is proportional to the square root of the oxygen concentration. Francis<sup>7,8)</sup> found the oxidation in permanganate to be of the first order. Schmidt and Elder<sup>9)</sup>, also using bituminous coals, found the rate to be proportional to the 0.61 power of the oxygen concentration.

In this section Scott's methods<sup>9)</sup> are checked by experiments with Oyubari coal with the conclusion that they appear to be appropriate.

Table 2 shows the results of experiments on oxidation at a constant temperature when the diameter of the glass pipe is 3.1 cm, the sectional area is  $7.54\text{ cm}^2$  and the average length of pieces in 100 grams of coal is 18.54 cm.



TABLE 2. Effect of time of contact on the oxidation rates of coal in air

| Test No. | Size mesh cm | Temper-ature | Pres-sure | Coal length | Rate          | Contact time | Effluent gas analysis, percent by volume |                |      |                | log (O <sub>2</sub> ) |
|----------|--------------|--------------|-----------|-------------|---------------|--------------|--|----------------|------|----------------|-----------------------|
|          |              | °C           | mmHg      | cm          | (N.T.P.) l/hr | sec          | CO <sub>2</sub>                          | O <sub>2</sub> | CO   | N <sub>2</sub> |                       |
| 1        | 20-23 (mesh) | 150          | 740       | 18.54       | 1.50          | 94.7         | 1.62                                     | 11.73          | 0.43 | 86.22          | 1.0693                |
| 1        |              | 150          | 738       | 18.54       | 2.15          | 65.6         | 0.83                                     | 14.48          | 0.25 | 84.44          | 1.1608                |
| 2        |              | 100          | 740       | 18.54       | 2.10          | 76.9         | 0.11                                     | 19.21          | 0.04 | 80.64          | 1.2835                |
| 3        |              | 50           | 746       | 18.54       | 0.9           | 209.0        | 0.04                                     | 19.92          | 0.00 | 80.04          | 1.2993                |
| 4        | 0.1 (cm)     | 150          | 740       | 18.54       | 1.4           | 101.6        | 0.26                                     | 18.58          | 0.09 | 81.07          | 1.2691                |
| 4        |              | 150          | 740       | 18.54       | 1.0           | 142.3        | 0.45                                     | 17.87          | 0.13 | 81.55          | 1.2521                |
| 4        |              | 150          | 740       | 37.08       | 1.0           | 284.6        | 0.76                                     | 15.76          | 0.26 | 83.22          | 1.1976                |
| 5        |              | 100          | 742       | 18.54       | 0.7           | 229.6        | 0.17                                     | 19.26          | 0.05 | 80.57          | 1.2847                |
| 5        |              | 100          | 744       | 18.54       | 0.6           | 268.8        | 0.19                                     | 18.98          | 0.05 | 80.78          | 1.2783                |
| 5        |              | 100          | 744       | 37.08       | 0.6           | 537.6        | 0.34                                     | 17.61          | 0.09 | 82.05          | 1.2458                |
| 6        |              | 50           | 742       | 18.54       | 0.8           | 233.0        | 0.01                                     | 20.44          | 0.00 | 79.55          | 1.3105                |
| 6        |              | 50           | 742       | 18.54       | 0.7           | 265.4        | 0.02                                     | 20.22          | 0.00 | 79.75          | 1.3058                |
| 6        |              | 50           | 742       | 37.08       | 0.7           | 530.8        | 0.03                                     | 19.57          | 0.00 | 80.40          | 1.2916                |
| 7        | 0.2 (cm)     | 150          | 740       | 18.54       | 1.0           | 142.3        | 0.38                                     | 18.92          | 0.11 | 80.70          | 1.2769                |
| 7        |              | 150          | 740       | 18.54       | 1.5           | 94.9         | 0.23                                     | 19.18          | 0.04 | 80.59          | 1.2829                |
| 7        |              | 150          | 740       | 37.08       | 1.5           | 189.8        | 0.41                                     | 18.22          | 0.13 | 81.24          | 1.2606                |
| 8        |              | 100          | 740       | 18.54       | 1.0           | 161.3        | 0.11                                     | 20.30          | 0.03 | 79.56          | 1.3075                |
| 8        |              | 100          | 742       | 18.54       | 0.7           | 229.3        | 0.12                                     | 19.58          | 0.04 | 80.26          | 1.2918                |
| 8        |              | 100          | 742       | 37.08       | 0.7           | 459.2        | 0.22                                     | 19.09          | 0.06 | 80.63          | 1.2808                |
| 9        |              | 50           | 744       | 18.54       | 0.6           | 309.9        | 0.01                                     | 20.67          | 0.00 | 79.32          | 1.3153                |
| 9        |              | 50           | 742       | 18.54       | 0.7           | 265.4        | 0.01                                     | 20.77          | 0.00 | 79.22          | 1.3174                |
| 9        |              | 50           | 742       | 37.08       | 0.7           | 530.8        | 0.04                                     | 20.57          | 0.00 | 79.39          | 1.3132                |

$$\text{Volume of voids} = \frac{7.54 \times 18.54 \times 0.45}{1000} = 0.0629 \quad 1$$

$$\text{Rate of circulation} = \frac{1000}{3600} = 0.000278 \text{ l/sec at N.T.P.}$$

$$1000 = \text{volume of air in l/hr}$$

$$3600 \text{ sec} \rightarrow 1 \text{ hour}$$

When the temperature of the electric furnace is 100°C at 740 mmHg,

$$0.000278 \times \frac{760}{740} \cdot \frac{373}{273} = 0.00039 \text{ l/sec}$$

$$\text{The time of contact is } \frac{0.0629}{0.00039} = 161.3 \text{ sec.}$$

In the same way the time of contact in the experiments is determined.

Here the correction of changes in volume by chemical reactions is not taken into consideration, because the approximate constancy of nitrogen concentration shown in the results of gas analysis in Table 2 indicates little necessity for correction and any errors in reaching a theoretical conclusion by experiment would be trivial.

Table 2 shows that the log of the shortage of oxygen concentration after the air has passed through the coal is inversely proportional to the time of contact with the coal. For example, in the test in which air was forced through fresh coal at 100°C, when the contact time was 230 seconds, the oxygen percentage in the effluent gases was 19.26 percent, when it was 269 seconds, the oxygen percentage was 18.98 and when it was 538 seconds, the oxygen percentage was 17.61.

When the results are applied to an examination of spontaneous combustion of coal in the underground, it is found that the percentage of oxygen concentration in effluent air has no value in connection with heat generation and its rate of increase. Provided that the speed of air passing through a body of broken coal is accelerated in accordance with the rise of temperature, the percentage of oxygen concentration in the effluent air increases, as the time duration of contact is a measurable factor. The percentage of oxygen concentration is low when the speed of air passing through the heat generation area is slow and the time duration of contact is long.

In Figure 4 is shown the relation between the time of contact and the log of oxygen concentration in the effluent air. The fact that the relation is shown in straight lines reveals that the reaction is primary and consequently the oxidation rate is proportional to the oxygen concentration. It was previously shown that,

$$-\frac{d[\text{O}_2]}{d\theta} = k[\text{O}_2] \quad (4)$$

$$\ln[\text{O}_2] = -k\theta + C$$

where  $C$  is a constant integral

when  $\theta = 0$ ,  $\text{O}_2 = 20.93$

and  $C$  is  $\log 20.93$  in the air.

$[\text{O}_2]$ : is the oxygen concentration in effluent air

$$\therefore [\text{O}_2] = 20.93 e^{-k\theta}$$

For any given time of contact  $\theta$ , one may write

$$[\text{O}_2]_{\text{ave}} \cdot d\theta = 20.93 e^{-k\theta} \cdot d\theta$$

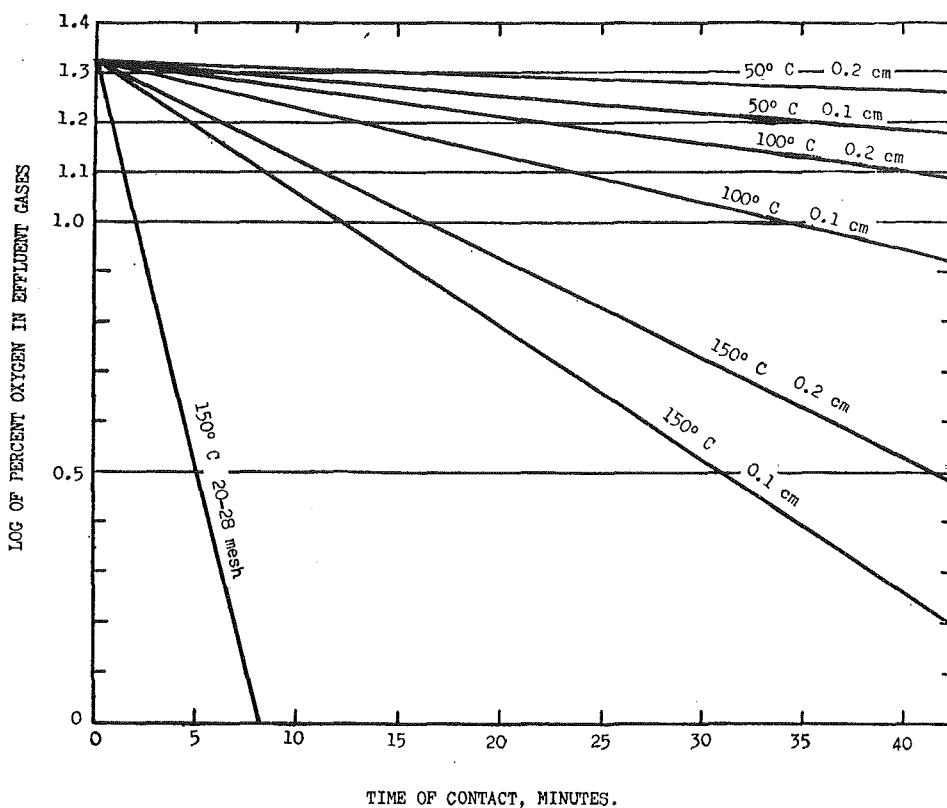


Fig. 4. Oxidation of coal; decrease in oxygen concentration with increase in contact time

which, when integrated and the proper limits are substituted gives:

$$\begin{aligned}
 [\text{O}_2]_{\text{ave}} \cdot \theta &= \frac{20.93 e^{-k\theta}}{-k} + \frac{20.93}{k} \\
 \text{or} \quad [\text{O}_2]_{\text{ave}} &= \frac{20.93 e^{-k\theta} - 20.93}{-k\theta} \\
 &= \frac{[\text{O}_2] - 20.93}{-k\theta} \\
 &= \frac{20.94 - [\text{O}_2]}{\ln \frac{20.93}{[\text{O}_2]}}
 \end{aligned}$$

This is a logarithmic mean. G. S. Scott takes pure air as a standard, and when the oxygen concentration of effluent air is not 20.93%, he proposes to

multiply the oxidation rate by the factor  $p$ . This factor  $p$  is

$$\text{factor } "p" = \frac{20.93 - [\text{O}_2]}{\ln \frac{20.93}{[\text{O}_2]}}$$

Figure 5 shows a plot of factor  $p$  against the effluent oxygen concentration and is much more convenient to use than the equation above. Examination of Figure 5 shows, for example, that when the oxygen concentration of effluent air is ten percent, the oxidation rate is 0.7 times as rapid as it would be if the effluent concentration were 20.93 percent.

The factor in the Schmidt method is also shown in Figure 5; the Winnill method is about midway between the two.

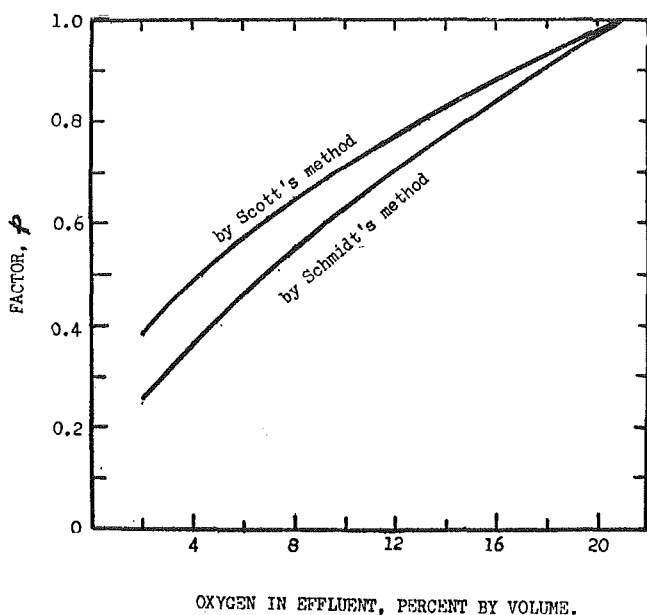


Fig. 5. Relation between effective oxygen concentration in contact with coal and oxygen concentration in effluent gases when entering gases contain 20.93 percent oxygen

## V. Work on the rate of temperature rise

If it is assumed that the particle diameter of coal, which has  $\Omega$  amount of oxygen, is  $d$  cm and the temperature is  $t^\circ\text{C}$ , in the case of perfect heat accumulation the oxidation rate is  $4\Omega$ , provided that the coal has absorbed  $4\Omega$  of oxygen in a given time. Consequently the amount of absorbed oxygen is  $\Omega + 4\Omega$ .

The rate of temperature rise caused by perfect heat accumulation is:

$$\frac{\text{calories per unit of oxygen consumed} \times \text{oxidation rate}}{\text{the weight of coal} \times \text{specific heat}}$$

As a matter of fact some calories are lost at the time of the inflow and outflow of gases, and are absorbed into the neighbouring rock by conduction, radiation and convection. Thus the following equation is necessary to indicate the rise of temperature in coal.

$$\text{Rate of temperature rise } \left( \frac{dt}{d\theta} \right) = \frac{\text{rate of heat generation} - \text{rate of heat dissipation}}{\text{quantities of materials} \times \text{specific heat}}$$

$$\begin{aligned} \text{Rate of heat generation } \left( \frac{dQ}{d\theta} \right)_g &= \text{Heat liberated per unit of oxygen consumed} \\ &\times \text{rate of oxidation } \left( \frac{d\Omega}{d\theta} \right) \\ &\quad (4.4 \text{ cal/cc of Oyubari low seam coal}) \end{aligned}$$

$$\begin{aligned} \text{Rate of heat dissipation } \left( \frac{dQ}{d\theta} \right)_L &= \text{Losses to circulating air and gases} \\ &+ \text{losses by conduction, radiation and convection} \end{aligned}$$

Herein the theory on the mechanism of an outbreak of spontaneous combustion of coal offered in the writer's preceding paper is more tangibly illustrated.

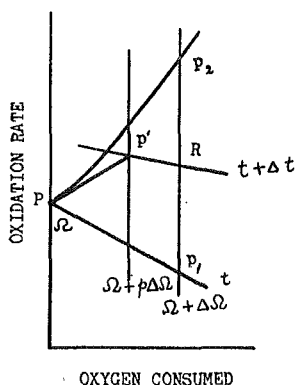


Fig. 6. Relation between oxidation rate and temperature rise, I.

If an optional point on  $P_1P_2$  is  $R$  and

$$\begin{aligned} P_1R &\propto (\text{the rate of heat generation} - \text{the rate of heat radiation}) \\ &\div (\text{the quantities of material} \times \text{specific heat}) = \Delta t \end{aligned}$$

In Figure 6, a perfect heat radiation curve and a perfect heat accumulation curve are drawn from an optional point  $P$ , when the temperature is  $t^\circ\text{C}$  and the amount of absorbed oxygen is  $\Omega$  cc. The curve of the oxidation rate per unit of time duration shows the amount of absorbed oxygen, and its points of intersection with the perfect heat radiation curve and the perfect heat accumulation curve are  $P_1$  and  $P_2$ .

Then:

$$P_1P_2 \propto (4.4 \times \Delta\Omega) \div (\text{quantities of materials} \times \text{specific heat})$$

where the line  $P_1P_2$  indicates the rate of temperature rise in the case of perfect heat accumulation.

$P_1R$  shows the actual rate of temperature rise.

A perfect heat radiation curve at the temperature of  $t + \Delta t$  is drawn through  $R$ . The factor of influence of the rate of oxygen concentration is " $p$ ", and the point of intersection with the line of the oxidation rate  $p\Delta\Omega$  and the above mentioned curve, is  $P'$ . Thus  $P'$  is a point on the practical heat accumulation curve which shows the change of the rate of temperature rise and the rate of oxidation during a given time.

Now  $P''$  is taken on the practical heat accumulation curve in the same way as in  $P'$ . Thus the practical curve of temperature rise can be determined.

Now this theory can be expanded. As Figure 7 shows,  $P'$  could be found outside of the  $P_1PP_2$  area because of giving off or receiving heat to or from other coal substances. The temperature rises at the back of the active oxidation zone in a coal oxidation path because of the transportation of radiated heat from the oxidation zone as well as from the heat produced by oxidation there. The heat radiated to the back is not very large. Thus  $R$  on the  $P_1P_2$  line comes closer to  $P_2$ : sometimes it is on an extension of the  $P_1P_2$  line.

As oxygen in the air is absorbed in the active oxidation zone, oxygen in low concentration oxidates the adjacent coal, thus  $p\Delta\Omega$  is smaller than  $\Delta\Omega$ .

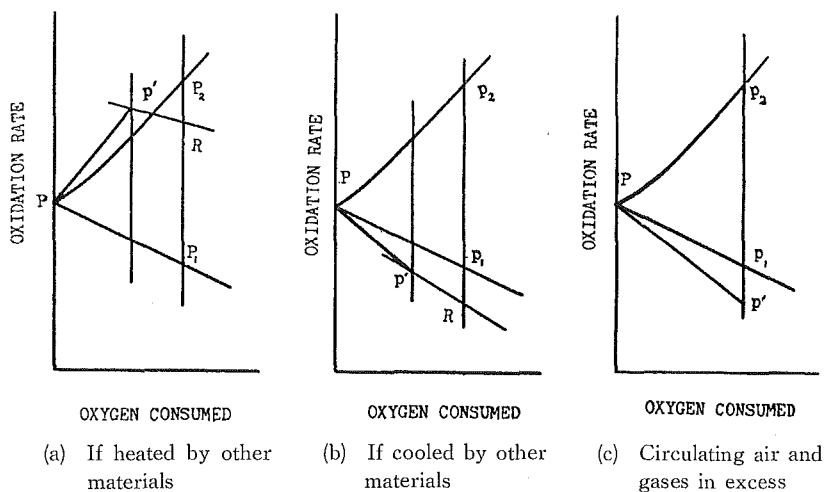


Fig. 7. Relation between oxidation rate and temperature rise II.

Consequently  $P'$ , which is the intersection point of the two curves, is located outside of the  $P_1PP_2$  area. In other words it could be said that there is coal with high oxidating potentiality at the back of any active oxidation zone.

$P'$  is also found outside of the  $P_1PP_2$  area when the heat generation rate

is less than the heat radiation rate. The temperature of this sort of outside coal is lowered by cooling and the oxidation rate declines rapidly.

## VI. Calculation of the practical rate of temperature rise

### (1) Temperature rise at normal temperatures

Consider fresh coal whose average particle diameter is 0.2 cm. The temperature of the coal and the atmosphere are assumed to be 25°C. As there is no difference of temperature between the coal and the atmosphere, there is no circulating air current—natural ventilation—, and no refrigeration by the atmosphere. Now the rate of the temperature rise of the coal is calculated when the heights of the coal piles are 2 meters, 4 meters and 6 meters.

When the sectional area of a coal column in a pile of coal is assumed to be 1 cm<sup>2</sup>, and the volume and the weight of the coal are measured and the oxidation rate and the amount of generated heat are given, the rate of temperature rise can be calculated.

When the height of the coal column is 2 meters, and the specific weight of coal is assumed to be 1.3 and the calculated fraction of voids in a pile of coal is assumed to be 0.45, the weight of 1 cm<sup>2</sup> of the coal column is

$$1.3 \times (1 - 0.45) \times 200 \times 1 \times 1 = 143 \text{ (g)}$$

$$\text{Volume of voids is } 200 \times 0.45 = 90 \text{ cc.}$$

$$\text{Volume of oxygen in the voids is } 90 \times 0.2093 = 18.8 \text{ cc.}$$

$$\text{The oxidation rate at 25°C is } 1.875 \text{ cc/hr. } 100 \text{ g.}$$

$$2.68 \text{ cc/hr. } 143 \text{ g.}$$

$$\text{The generated calories are } 4.4 \text{ cal/cc, and } 11.8 \text{ cal/hr. } 143 \text{ g.}$$

As the temperature of the coal and the atmosphere are 25°C, there is no refrigeration from the outside, and no loss of the generated heat. Oxidation is possible here for the first hour, because the volume of oxygen in the voids is 18.8 cc and the absorbed amount of oxygen is 2.68 cc.

Therefore the actual rate of average temperature rise is

$$\frac{11.8}{0.25 \times 143} = 0.33 \text{ °C/hr.}$$

In the same way it is 0.33 °C/hr. at the height of both 4 meters and 6 meters.

This oxidation rate is applicable in the first hour, but the amount of oxygen in the voids is only 18.8 cc, which is so small as to allow a temperature rise of only a few degrees. Thus the circulating air current caused by temperature rise should be considered.

Since the eventual temperature rise is of only a few degrees, the circulating air current is very slight. Thus whether enough air to continue the oxidation can be supplied is questionable; it is impossible when the diameter of the coal particles is smaller.

In fresh coal, methane gas is produced. Though the relation between the mechanism of methane production and the absorption of oxygen is not clear now, it is known that, when coal is broken, more methane will be produced and oxidation will become more active as the new surfaces are exposed. This phenomenon, however, declines as time passes.

The air supply resulting from the methane production in coal increases rapidly till the methane concentration rate reaches 30%; the ascending air current reaches a maximum when it is 30–40%. Therefore at the very beginning of a temperature rise in coal, methane production makes the spontaneous generation of combustion easier by drawing in more oxygen from the atmosphere.

## (2) Temperature rise in low temperature oxidation

The oxidation rate of coal at 50°C is 2.1 cc/hr. 100 g in the course of perfect heat accumulation. Usually, however, it is not 2.1 cc/hr. 100 g because

TABLE 3. Relation between the temperature of coal and the amount of heat accumulation

|   |                         | temperature of coal 50°C<br>average particle diameter 0.2 cm   |       |       |
|---|-------------------------|--|-------|-------|
| Ascending air current                     |                         | 6.9 l/day·1 cm <sup>2</sup> , 287.5 cc/hr. 1cm <sup>2</sup>  |       |       |
| Velocity of air current                   |                         | $\frac{6900}{0.45 \times 1 \times 1} = 153000 \text{ cm/day,}$<br>634 cm/hr, 10.6 cm/min.                            |       |       |
| Height of coal column                     | m                       | 2  | 4     | 6     |
| Weight of coal column per/cm <sup>2</sup> | g                       | 143  | 286   | 429   |
| Oxidation rate at 50°C                    | cc/hr                   | 3.0  | 6.0   | 9.0   |
| Generated calories                        | cal/hr                  | 13.2   | 26.4  | 39.6  |
| Contact time                              | min                     | 18.8   | 37.6  | 56.3  |
| Volume of oxygen in the voids             | cc/hr. 1cm <sup>2</sup> | 60.17  | 60.17 | 60.17 |
| Percentage of absorbed oxygen             | %                       | 1.04   | 2.09  | 3.13  |
| Oxygen percentage in effluent gases       | %                       | 19.89  | 18.84 | 17.80 |
| Rate of heat dissipation                  |                         | $\frac{6.9}{22.4} \times 6.9 \times 25 = 53.14 \text{ cal/day} \cdot 1 \text{ cm}^2,$<br>2.2 cal/hr·1cm <sup>2</sup> |       |       |
| Actual amount of heat accumulation        | cal/hr                  | 11.0   | 24.2  | 37.4  |
| Actual rate of average temperature rise.  | °C/hr                   | 0.31   | 0.34  | 0.35  |



the coal is refrigerated by circulating air currents and because of difference of temperature from that of the environment, which facts, tend to cause lowering of the temperature down to 50°C. For the present calculation, however, the above numerical value of oxidation rate is used.

The heights of the coal column here likewise are 2, 4 and 6 meters. The oxidation rate changes according to the duration of contact. The log of the degree of oxygen concentration in effluent gas and the length of time of contact are conversely proportionate. Therefore if the degree of oxygen concentration is calculated by multiplying the coefficient "*p*" by the oxidation rate, the oxidation rate for any given length of time of contact can be known. As the degree of oxygen concentration in effluent air is high, the coefficient one is

TABLE 4. Relation between the particle diameter  
temperature of atmosphere 25°C.

|   |                            |   |       |       |
|---|----------------------------|---|-------|-------|
| Average particle diameter                   | cm                         | 1   |       |       |
| Temperature of atmosphere                   | °C                         | 25  |       |       |
| Temperature of coal                         | "                          | 50  |       |       |
| Ascending air current                       |                            | $K_s = 76.8 \text{ l/day} \cdot 1 \text{ cm}^2$<br>3200 cc/hr · 1 cm <sup>2</sup>                                   |       |       |
| Calculated fraction of Voids                |                            | $V = 0.45$  |       |       |
| Velocity of air current                     |                            | 170,670 cm/day<br>7,110 cm/hr<br>118.5 cm/min   |       |       |
| Height of coal column                       | m                          | 2   | 4     | 6     |
| Contact time                                | min                        | 1.7   | 3.4   | 5.1   |
| Weight of coal column per 1 cm <sup>2</sup> | g/cm <sup>2</sup>          | 143   | 286   | 429   |
| Oxidation rate at 50°C                      | cc/hr · 1 cm <sup>2</sup>  | 0.63  | 1.26  | 1.89  |
| Rate of heat generation                     | cal/hr · 1 cm <sup>2</sup> | 2.8   | 5.5   | 8.3   |
| Volume of oxygen in air current             | cc/hr · 1 cm <sup>2</sup>  | 670   |       |       |
| Percentage of absorbed oxygen               | %                          | 0.02  | 0.04  | 0.06  |
| Oxygen percentage in effluent gases         | %                          | 20.91   | 20.89 | 20.87 |
| Rate of heat dissipation                    |                            | $\frac{76.8}{22.4} \times 6.9 \times 25 =$<br>591.4 cal/day · 1 cm <sup>2</sup><br>24.64 cal/hr · 1 cm <sup>2</sup> |       |       |
| Actual amount of heat accumulation          | cal/hr                     | No heat<br>accumulation   |       |       |
| Actual rate of average temperature rise     | °C/hr                      |   |       |       |

used in the calculations here. (Table 3)

To make the problem simple the oxidation rate is calculated in the case of perfect heat accumulation. As was pointed out before, the actual oxidation rate is lower than 2.1 cc/hr. 100 g and the rate of temperature rise is usually lower than expected. The results of this calculation show that the amount of the residual coal does not have a very serious influence upon the rate of temperature rise at the beginning.

Table 4 shows the relation between the change of the particle diameter of coal and the amount of heat accumulation.

According to Table 4 temperature rise does not occur when the particle diameters are 1 cm and 0.5 cm, but there is consequent heat accumulation when

and the amount of heat accumulation

temperature of coal 50°C.

|  |       |       |  |       |       |  |       |       |                             |                        |  |
|--|-------|-------|--|-------|-------|--|-------|-------|-----------------------------|------------------------|--|
| 0.5  |       |       | 0.2  |       |       | 0.1  |       |       | Remarks                     |                        |  |
| 25   |       |       | 25   |       |       | 25   |       |       |                             |                        |  |
| 50   |       |       | 50   |       |       | 50   |       |       |                             |                        |  |
| 37.1 l/day·1 cm <sup>2</sup><br>1546 cc/hr·1 cm <sup>2</sup>     |       |       | 6.9 l/day·1 cm <sup>2</sup><br>287.5 cc/hr·1 cm <sup>2</sup> |       |       | 0.58 l/day·1 cm <sup>2</sup><br>24.0 cc/hr·1 cm <sup>2</sup> |       |       |                             |                        |  |
| 0.45   |       |       | 0.45   |       |       | 0.45   |       |       |                             |                        |  |
| 82,400 cm/day<br>3,400 cm/hr<br>57.3 cm/min                      |       |       | 15,300 cm/day<br>638 cm/hr<br>10.64 cm/min                   |       |       | 1,280 cm/day<br>53.4 cm/hr<br>0.89 cm/min                    |       |       |                             |                        |  |
| 2  | 4     | 6     | 2  | 4     | 6     | 2  | 4     | 6     |                             |                        |  |
| 3.5  | 7.0   | 10.5  | 18.8   | 37.6  | 56.3  | 224.7  | 449.4 | 674.2 |                             |                        |  |
| 143  | 286   | 429   | 143  | 286   | 429   | 143  | 286   | 429   | Specific weight of coal 1.3 |                        |  |
| 1.23   | 2.46  | 3.69  | 3.00   | 6.00  | 9.01  | 6.15   | 12.30 | 18.4  | Particle diameter           | Oxidation rate at 50°C |  |
| 5.41   | 10.82 | 16.23 | 13.2   | 26.4  | 39.6  | 22.0   |       |       |                             | 1 cm 0.44 cc/hr·100 g  |  |
|  |       |       |  |       |       |  |       |       |                             | 0.5 cm 0.86 "          |  |
|  |       |       |  |       |       |  |       |       |                             | 0.2 cm 2.1 "           |  |
|  |       |       |  |       |       |  |       |       | 0.1 cm 4.2 "                |                        |  |
| 323.5  |       |       | 60.17  |       |       | 5.0  |       |       |                             |                        |  |
| 0.80   | 1.49  | 2.29  | 1.04   | 2.09  | 3.13  | 20.93  |       |       |                             |                        |  |
| 20.13  | 19.44 | 18.64 | 19.89  | 18.84 | 17.80 | 0  |       |       | Coefficient $p=1$           |                        |  |
| 285.7 cal/day·1 cm <sup>2</sup><br>11.9 cal/hr·1 cm <sup>2</sup> |       |       | 53.1 cal/day·1 cm <sup>2</sup><br>2.2 cal/hr·cm <sup>2</sup> |       |       | 4.6 cal/day·cm <sup>2</sup><br>0.19 cal/hr·1 cm <sup>2</sup> |       |       |                             |                        |  |
| No heat accumulation   |       | 4.33  | 11.0   | 24.2  | 37.4  | 21.81  |       |       |                             | * Arithmetic average   |  |
|  |       | 0.04  | 0.31   | 0.34  | 0.35  | 0.61   |       |       |                             |                        |  |

TABLE 5. Relation between the temperature and average particle diameter 0.2 cm

| Temperature of coal   | °C                       | 25                             |      |      |
|---|--------------------------|--------------------------------|------|------|
| Ascending air current                                       |                          | 0                              |      |      |
| Calculated fraction of voids                                |                          | 0.45                           |      |      |
| Velocity of air current                                     |                          | 0                              |      |      |
| Height of coal column                                       | m                        | 2                              | 4    | 6    |
| Contact time  | min                      | $\infty$                       |      |      |
| Weight of coal column per 1 cm <sup>2</sup>                 | g/cm <sup>2</sup>        | 143                            | 286  | 429  |
| Log of the degree of oxygen concentration in effluent gases |                          |                                |      |      |
| Oxygen concentration in effluent air                        | %                        |                                |      |      |
| Coefficient $p$   |                          |                                |      |      |
| Oxidation rate  | cc/hr·1 cm <sup>2</sup>  | 2.68                           | 5.36 | 8.04 |
| Rate of heat generation 4.4 cal/cm <sup>3</sup>             | cal/hr·1 cm <sup>2</sup> | 11.8                           | 23.6 | 35.4 |
| Rate of heat dissipation                                    |                          | 0                              |      |      |
| Actual amount of heat accumulation                          | cal/hr                   | 11.8                           | 23.6 | 35.4 |
| Actual rate of average temperature rise                     | °C/hr                    | $\frac{11.8}{0.25} \times 143$ |      |      |
|   |                          | 0.33                           | 0.33 | 0.33 |

|                                     |                         |      |      |      |
|-------------------------------------|-------------------------|------|------|------|
| Another Solution (1)                |                         |      |      |      |
| Temperature of coal                 | °C                      | 25   |      |      |
| Volume of oxygen in air current     | cc/hr·1 cm <sup>2</sup> | 18.8 | 37.7 | 56.5 |
| Oxygen percentage in effluent gases | %                       |      |      |      |
| Percentage of absorbed oxygen       | %                       |      |      |      |
| Volume of absorbed oxygen           | cc/hr·1 cm <sup>2</sup> |      |      |      |

|                                     |                         |                            |  |  |
|-------------------------------------|-------------------------|----------------------------|--|--|
| Another Solution (2)                |                         | Schmidt's coefficient $p'$ |  |  |
| Oxygen percentage in effluent gases | %                       |                            |  |  |
| Coefficient $p'$                    |                         |                            |  |  |
| Oxidation rate                      | cc/hr·1 cm <sup>2</sup> |                            |  |  |

they are 0.2 cm and 0.1 cm.

When the particle diameters are 0.5 cm and 0.2 cm, and the circulating air current is plentiful, the deeper back portion of the current has the higher temperature, but when the particle diameter is 0.1 cm and circulating air current is small, oxidation is most active at the intake side.

Though heat loss is more or less considered in this section, the effect of

the amount of heat accumulation  
temperature of atmosphere 25°C

| 50   | 100   | 150   | Remarks  |
|--|---|---|--|
| 6.9 l/day·1 cm <sup>2</sup><br>287.5 cc/hr·cm <sup>2</sup><br>0.45 | 10.6 l/day·1 cm <sup>2</sup><br>441.7 cc/hr·1 cm <sup>2</sup><br>0.45 | 11.3 l/day·1 cm <sup>2</sup><br>470.8 cc/hr·1 cm <sup>2</sup><br>0.45 | Oxidation rate<br>at 25°C<br>1.875cc/hr. 100 g<br>at 50°C<br>2.1 cc/hr. 100 g<br>at 100°C<br>9.1 cc/hr. 100 g<br>at 150°C<br>40 cc/hr. 100 g |
| 15300cm/day, 638cm/hr<br>10.6cm/min<br>2      4      6             | 23600cm/day, 981cm/hr<br>16.4cm/min<br>2      4      6                | 25100cm/day, 1046cm/hr<br>17.4cm/min<br>2      4      6               |  |
| 18.8    37.6    56.3   | 12.2    24.2    36.7  | 11.5    22.9    34.4  |  |
| 143    286    429  | 143    286    429   | 143    286    429   |  |
| 1.29   1.260   1.235   | 1.26   1.19   1.14  | 1.12   0.85   0.637   |  |
| 19.5   18.2   17.2   | 18.2   15.5   13.8  | 13.2   7.08   4.33  |  |
| 0.98   0.95   0.92   | 0.95   0.88   0.81  | 0.80   0.62   0.50  |  |
| $2.1 \times 0.98 \times \frac{143}{100}$                           | $0.1 \times 0.95 \times \frac{143}{100}$                              | $40 \times 0.8 \times \frac{143}{100}$                                |  |
| 2.95   5.71   8.28   | 12.7   22.9   31.5  | 45.7   71.1   85.8  |  |
| 13.0   25.1   36.4   | 55.9   100.8   138.6  | 200.1   312.8   377.5   |  |
| 53.1 cal/day·1 cm <sup>2</sup><br>2.2 cal/hr·1 cm <sup>2</sup>     | 244.9 cal/day·1 cm <sup>2</sup><br>10.2 cal/hr·1 cm <sup>2</sup>      | 535.1 cal/day·1 cm <sup>2</sup><br>18.1 cal/hr·1 cm <sup>2</sup>      |  |
| 10.8   22.9   34.2   | 45.7   90.6   128.4   | 182.0   294.7   359.4   |  |
| 0.30   0.32   0.32   | 1.28   1.27   1.20  | 5.10   4.12   3.35  |  |

| 50                  | 100                 | 150                 | Oxidation rate |
|---------------------|---------------------|---------------------|----------------|
| 60.17               | 92.45               | 98.5                |                |
| 19.5   18.2   17.2  | 18.2   15.5   13.8  | 13.2   7.08   4.33  |                |
| 1.4    2.7    3.7   | 2.7    5.4    7.1   | 7.7   13.85   16.60 |                |
| 4.03   7.75   10.30 | 11.95   23.5   31.4 | 36.3   65.3   78.1  |                |

|                    |                    |                    |  |
|--------------------|--------------------|--------------------|--|
| 19.5   18.2   17.2 | 18.2   15.5   13.8 | 13.2   7.08   4.33 |  |
| 0.97   0.94   0.90 | 0.94   0.82   0.77 | 0.76   0.54   0.40 |  |
| 2.91   5.66   8.10 | 12.2   21.3   30.0 | 43.5   62.0   69.0 |  |

heat transportation in coal columns and the deterioration of oxygen concentration is not discussed. If such points were considered, the calculations would become considerably more complicated. Only when the O<sub>2</sub> concentration in effluent gasses is 0% with 0.1 cm particle diameter, is the full consideration of the effect of the deterioration of oxyge concentration necessary.

### (3) Temperature rise in high temperature oxidation

Table 5 shows the relation between the temperature change from 25°C to 150°C and the amount of heat accumulation when the particle diameter of coal is 0.2 cm.

In this section the effect of the change in oxygen concentration upon the oxidation rate is considered. As the log of the degree of oxygen concentration and the contact time change from a straight line, the log of the degree of oxygen concentration in effluent gases with the contact time, and the actual oxidation rate are obtained by multiplying the oxidation rate by the coefficient  $p$ .

Next, from the degree of oxygen concentration in effluent air, the percentage of absorbed oxygen can be known. From this value together with absorption from the circulating air current, the volume of absorbed oxygen is computed. Then the volume of absorbed oxygen within a unit of time is compared with the actual oxidation rate which has been discussed in the preceding section.

According to Schmidt and Elder the oxidation rate is proportional to the 0.61 power of the oxygen concentration. Consequently the actual oxidation rate is computed by obtaining the coefficient " $p$ " of the oxidation rate.

The oxidation rate can be computed by the three methods mentioned above. Such computations were made as displayed in Table 5; it is found that most of the results have errors of less than 10%.

If the temperature rises higher and oxidation becomes active, the oxidation phenomena are more complicated.

When the temperature is higher than 150°C, the ascending air current tends to decrease. Therefore there is oxidation with an oxygen shortage at the return side, though the oxidation is active at the intake side, which is the high temperature zone. Temperature rise is caused not only by oxidation but also by convection of the high temperature effluent gases. The greater the distance from the active oxidation zone, the less the effect of heat transfer by convection.

This tendency is stronger in a coal mine with gradually inclined seams. When the excavation height is about 2-3 meters, and the length between the intake road and the return way, in long-wall mining method, is more than 100 meters, the heightened temperature in an entire gob in low-temperature oxidation with little leakage of air does not rise significantly near the return side, while the temperature rise is remarkable at the intake side. Consequently sometimes near the return side the tentative temperature rise is counteracted by a gradual cooling effect. This has been demonstrated by actual underground measurements at Yūbari coal mine in Hokkaido.

Generally speaking remarkable phenomena of high temperature heat generation by oxidation are in the following forms: (a) the transfer of the heat

generation zone toward the intake side, and (b) the extreme development of coal with a high oxidation potential.

(a) The transfer of the heat generation zone toward the intake side.

The causes of the transfer of the heat generation zone toward the intake side are as follows:

- i) in flow and outflow of air due to changes of atmospheric pressure,
- ii) changes of the resistance to air flow in a roadway,
- iii) decrease of circulating air currents at high temperatures,
- iv) differences in the directions of air leakage caused by the ventilating pressure in the underground and of ascending air flow caused by natural ventilation, particularly in the cases of flat seams,
- v) breathing air of heat generating coal in the process of oxidation,

Low-temperature fresh coal close to a high-temperature zone has high possibilities of temperature rise.

Consider fresh coal at 25°C with 0.1 cm particle diameter and oxidated coal which has become 100°C hot from 25°C through heat generation by oxidation. If the coal of 100°C is assumed to have a volume of 600 cc/100 g of absorbed oxygen, the oxidation rate of the coal is 15 cc/hr. 100 g.

Now suppose that the comparative contact times of the high temperature gases of 100°C and the inflow air of 25°C to the fresh coal of 25°C with a 0.1 cm particle diameter are fifty-fifty. When the coal of 25°C comes in contact with the high temperature gases of 100°C, the coal has an oxidation rate of 30 cc/hr. 100g, and when the coal comes in contact with the inflow air of 25°C, the coal has an oxidation rate of 3.75 cc/hr. 100g. The average oxidation rate is 16.9 cc/hr. 100g. This shows that the heat generated by oxidation is greater than that of the coal which has a volume of absorbed oxygen of 600 cc at 100°C.

Therefore not only the leveling of the high temperature zones is seen in the return side, but also there is a strong possibility of the transfer of heat generation zone by oxidation toward the intake side. The change is transitional.

(b) The development of coal with a high oxidation potential.

In the return side of a high temperature zone, sometimes the temperature rises over the perfect heat accumulation curve because of the conduction of heat and oxidation with the shortage of oxygen. If the coal in this status is supplied with fresh air, it will generate heat by oxidation very actively. This sort of coal is called "coal with a high oxidation potential."

Here is the danger of the "digging out coal method," since by this method the possibility of an outbreak of spontaneous combustion is enhanced. There

lies a zone with a high oxidation potential at the back of high temperature zone caused by heat generation, but it is impossible to discriminate this coal from the other. The removal of high temperature coal by digging out, means an increased oxygen supply to the neighbouring coal with a high oxidation potential. Therefore the temperature is not so high immediately after digging out, but there is danger of a rapid temperature rise.

Sometimes the temperature rises rapidly because of the changes of ventilation in the underground, which is one of the most serious causes of spontaneous combustion of coal. There has been no other explanation of this problem of spontaneous combustion than unpredictable leakage, and that caused by heat generation by oxidation. According to this present theory, it could be added that the supply of fresh air to the zone with a high oxidation potential which is located at the back of heat generation zone is also one of the causes of spontaneous combustion.

## VII. Conclusion

In this paper causes of an outbreak of spontaneous combustion of coal have been quantitatively discussed according to their importance and in order from oxidation at normal temperature to oxidation at high temperatures. In other words, necessary ventilation for heat generation by oxidation, the consequent heat conduction and radiation, and oxidation control by decrease of oxygen concentration resulting from oxidation are discussed in order. The basic theory here, however, is just an expansion of the theory discussed in the preceding paper.

It is possible to apply this theory to heat generation phenomena in the underground works without any contradictions. It must be noticed that a spontaneous generation of combustion in the underground is discussed in consideration of spontaneous generation of combustion of storage coal.

This theory will provide answers to the problems regarding every case of spontaneous generation of combustion of coal such as the cause of spontaneous generation of combustion at the actual spots, heat generation phenomena by oxidation, their control and preventive measures, etc.

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