Evaluation of Spontaneous Combustion in Stockpile of Sub-bituminous Coal

Dr. Haeyang PAK [*1], Toshiya TADA [*2], Naoki KIKUCHI [*3], Takuo SHIGEHISA [*3], Toru HIGUCHI [*3], Dr. Seiichi YAMAMOTO [*4]

[*1] Mechanical Engineering Research Laboratory, Technical Development Group
[*2] Mechanical Engineering Research Laboratory, Technical Development Group (currently Kobelco Eco-Solutions Co., Ltd.)
[*3] Coal & Energy Technology Dept., Technical Development Group
[*4] Coal & Energy Technology Dept., Technical Development Group (currently New Energy and Industrial Technology Development Organization)

Spontaneous combustion in coal stockpiles is one of the problems encountered when utilizing coals such as sub-bituminous coal and lignite that contain highly volatile matter. A method has been developed for simulating the spontaneous combustion in coal stockpiles. This method involves unsteady analysis taking into account the flow behavior of air flowing through the pile, low-temperature oxidation behavior of coal in the pile, evaporation, absorption and desorption behaviors of moisture in the coal. The simulation enables the evaluation of the temperature change in a coal stockpile without any large-scale temperature measurement. The simulation results show that the heat tends to be generated at the foot of each stockpile where the breathability is high. It has also been confirmed that the stockpile of sub-bituminous coal exhibits a faster temperature rise because the coal has an oxidation reactivity higher than that of bituminous coal.

Introduction

Coal is the resource that accounts for approximately 24% of the primary energy consumed in Japan, which imports most of this resource from other countries. According to Trade Statistics of Japan, the amount of coal imported reached 185.15 million tonnes in 2012.[1)] Approximately 60 percent of this amount is occupied by steaming coal used for fueling commercial and industrial boilers. In many cases, high-grade bituminous coal is used for this purpose. A coal with a higher degree of coalification has a higher carbon content. The Japanese Industrial Standard (JIS M1002) classifies coals according to their calorific values on the moisture-ash-free basis: i.e., bituminous coal with a calorific value of 33,910kJ/kg or higher, sub-bituminous coal with a calorific value from 30,560kJ/kg to 33,910kJ/kg (exclusive of 33,910kJ/kg), and lignite (brown coal) with a calorific value from 24,280kJ/kg to 30,560kJ/kg (exclusive of 30,560kJ/kg).

Coal-fired power generation plants are equipped with facilities for storing coal imported from other countries. These storage facilities are roughly classified into two types; i.e., outdoor coal storage and indoor coal storage. In outdoor coal storage such as that involving a stockpile (hereinafter "pile"), stackers and reclaimers are used for piling and delivering the coal; therefore, this type of storage is used in many cases by steelworks and thermal power generation plants on large sites. On the other hand, indoor coal storage, using silos, for example, is increasingly being adopted these days by plants with limited space and/or those with concern for the neighborhood environment.[2)] A coal yard generally has a storage capacity equivalent to the amount of coal consumed in one to two months, depending on the scale of the steelworks or thermal power generation plant.

When storing coal for an extended period of time, attention must be paid to the possibility of spontaneous combustion. The temperature inside a pile immediately after the piling is approximately 30℃ to 40℃, but it rises gradually due to the heat generated by low-temperature oxidation of the coal. The temperature of a pile is determined by the balance of the heat of coal oxidation, the latent heat of water evaporation and the heat dissipated from the pile by air flow. It is considered that the temperature continues to rise at a spot where heat generation dominates, which eventually leads to spontaneous combustion.[3)]

In many cases, the spontaneous combustion characteristics of coal are evaluated by measuring the change of temperature caused by the self-heating of a sample in an adiabatic system.[4)] It has been known that coal with strong oxidizing properties, such as a high O/C ratio (the ratio of oxygen to carbon contained in coal) and large specific surface area, is more prone to spontaneous combustion (internal factors). When considering the spontaneous combustion characteristics of coal in storage, the effect of external factors, such as the particle size distribution and the filling state of the coal, the amount of sprinkled/precipitated water and ambient temperature, must also be taken into account. As an evaluation method taking into consideration both the internal and external factors, long-term experiments extending over several months have been conducted to measure the temperature of test piles, each consisting of several thousand tonnes of coal.[5), 6)] This method is useful for evaluating the spontaneous combustion characteristics of a new type of coal that has not been used before, but is costly.

Recently, the supply sources of coal are becoming more diversified and coals are becoming more degraded. As a result, thermal power generation plants in Japan are using various types of coal. Above all, these plants are increasingly using coals such as sub-bituminous coal, which has a high O/C ratio and is more prone to spontaneous combustion than bituminous coal. These types of coals are expected to see increased use. Therefore, it is important to understand the spontaneous combustion characteristics of new types of coals in advance so as to prevent them from causing fires at coal yards.

It was against this background that Kobe Steel has developed a simulation technology for predicting the heat generation behavior inside a coal pile, taking into account internal factors such as the low-temperature oxidation of coal and external factors such as piling conditions and the outer environment, so as to prevent fires from occurring when storing a new type of coal, including sub-bituminous coal, that has not been used before.

1. Simulation model outline

A heat-generation simulation of a coal pile (hereinafter "pile simulation") has been designed to accurately reproduce the heat generation behavior inside piles. The following three behaviors were studied in detail and were modeled:

i) the behavior of air flowing inside the pile,
ii) the low-temperature oxidation behavior of coal inside the pile, and
iii) the evaporation and adsorption/desorption behaviors of moisture in the coal.

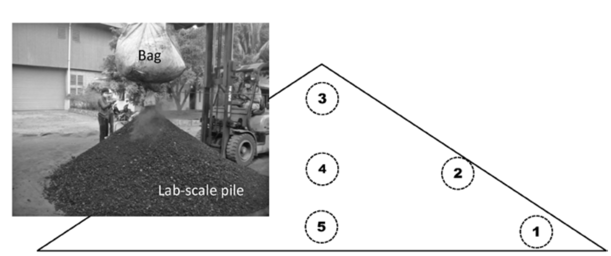
These models were incorporated into the software for general-purpose thermal fluid analysis (ANSYS Fluent), which makes it possible to perform unsteady analysis taking into account the heat transfer, flow and reaction in a large-scale pile. As a result, the heat generation characteristics of various coals during storage can now be predicted by simply obtaining their physical properties from small-scale tests. This eliminates the need for measuring the temperatures of large test piles.

1.1 Air flow behavior inside piles

In a pile, air flows by the natural convection associated with heat generation. Roughly speaking, the air flow is caused by warmed air rising up inside the pile and escaping outside, while fresh air is being supplied from outside. A pile is an accumulation of coal granules having a size distribution. Thus, in order to reproduce the air flow, the pressure drop inside the pile must be assessed and the flow conditions of the air must be determined. In addition, a pile consists of coal granules with a wide size distribution, resulting in different degrees of breathability, depending on the location within the pile. For example, the breathability is increased at the foot of a pile, because, when coal is piled by a stacker, coal granules with larger sizes tend to roll and to be segregated at the foot.

In order to evaluate the air flow distribution inside a pile, the granule size distributions were measured at various locations within a pile and the pressure drop for each granule size distribution was determined. **Fig. 1** shows the outline of a 1.5m laboratory pile simulating a stacking process. As shown in this figure, the test includes piling coal by discharging it from a container bag above the piling spot and collecting coal granules from areas spaced at a height interval of 0.5m. The coal granules thus collected were measured to determine their granule size distribution and coefficient of air flow resistance, as described later. The air flow resistance coefficient is used as a measure of breathability. This figure also includes average granule sizes (50% of integrated value) in different areas. Area 1 corresponds to the foot of the pile and shows an average granule size of 29.8mm, indicating that granules larger than others by one digit tend to accumulate there.

Next, an apparatus for measuring pressure drop, as shown in **Fig. 2**, was used to determine the relationship between the flow velocity and pressure drop in each area. This apparatus comprises a column to be packed with granules of coal, an air supply, a flow meter and a manometer for measuring the differential pressure of the packed column. **Fig. 3** depicts the relationships between the flow rate and pressure drop in areas 1 to 3. These areas correspond to the surface layer of the pile. This figure shows that, regardless of the area, a linear relationship as expressed by Equation (1) is established between the flow rate (u) and pressure



Particle diameter (50% of Integrated value)
Area 1: 29.8 mm Area 2: 6.2 mm Area 3: 6.3 mm
Area 4: 4.2 mm Area 5: 2.2 mm

Fig. 1 Piling test

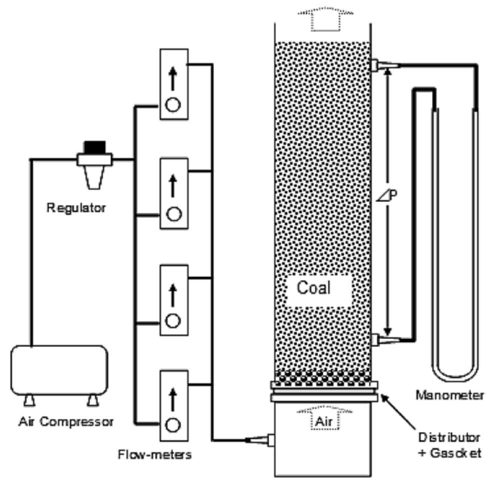


Fig. 2 Schematics of experimental apparatus for pressure drop

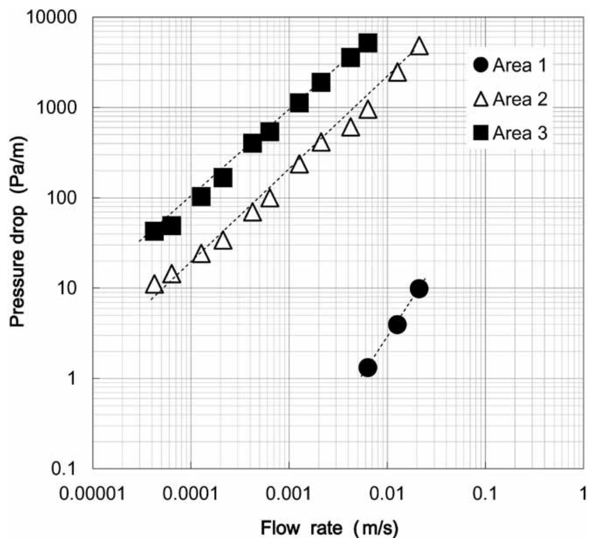


Fig. 3 Relationship between pressure drop and flow rate

drop (ΔP):

$$\Delta P/L = k \cdot u \quad \cdots\cdots (1)$$

wherein L is the height of the packed layer, while k is the slope of Equation (1) and represents the air flow resistance coefficient ($Pa \cdot s/m^2$). In this simulation, each pile is provided with a distribution of air flow resistance coefficient, k, so that the pressure drop can be determined and the behavior of the air flow inside the pile can be expressed. Considered by area, the air flow resistance coefficient becomes smaller at the foot (Area 1) of the pile, the area where larger granules tend to accumulate. For a conical shaped pile, the air flow resistance coefficient is higher in the areas deeper inside the pile and in the areas closer to the top.

1.2 Low-temperature oxidation behavior of coal inside pile

It is generally understood that, when coal is oxidized at a low temperature, the adsorption of oxygen leads to the production of peroxides first, and these peroxides decompose into CO, CO_2 and H_2O in the end. It is known that, as the integrated amount of oxygen adsorbed on the coal increases, the number of adsorption sites decreases, lowering the rate of adsorption. This means that, in a pile, the heat generating reaction proceeds by consuming oxygen in the air and, as time passes, the activity of this reaction decreases, slowing down the heat generation.

As described above, the heat is considered to be generated by the progress of a low-temperature oxidation reaction. In this simulation, Equation (2) is used to determine the heat-generation rate Q (kJ/m^3/s) of coal:

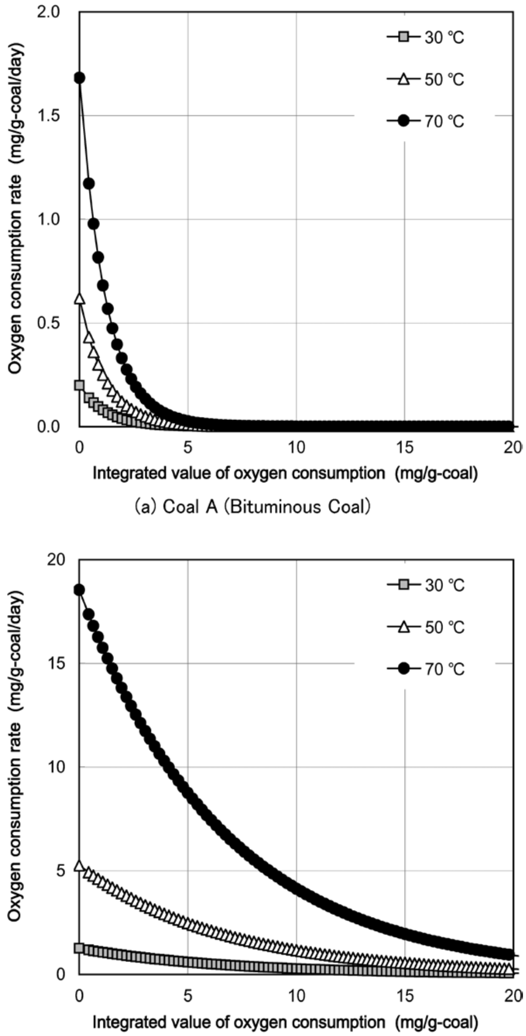
$$Q = \Delta H\,(1-\varepsilon)\,\rho\,OCR \quad \cdots\cdots (2)$$

wherein ΔH is the heat of oxidation reaction (kJ/kg-O_2), ε is the void ratio inside the pile and ρ is the solid density (kg/m^3). An important factor in determining the heat generation rate is the oxygen consumption rate (OCR) expressed in mg-O_2/g-coal/day. The OCR is a value unique to each coal and, as shown by Equation (2), a larger OCR indicates that more heat is generated in the pile. Furthermore, the OCR depends on the temperature and oxygen concentration. Thus, it is expressed by Equation (3) in this simulation.

$$OCR = OCR_0\,\mathrm{EXP}\{-\Delta E/\mathrm{R}(1/T - 1/T_0)\}[O_2]^n \quad \cdots\cdots (3)$$

wherein OCR_0 is a value (mg-O_2/g-coal/d) measured at 303K (30℃) with 21%-O_2 (standard conditions), ΔE is the activation energy (kJ/mol), $[O_2]$ is the oxygen concentration ratio (= "oxygen concentration in the pile" / "oxygen concentration under the standard conditions"), n is the order of reaction (-), and T and T_0 are the temperature (K) inside the pile and the temperature (K) under standard conditions, respectively. Here, the OCR_0 decreases with the increase in the integrated amount of oxygen consumption and is expressed as a function of the integrated amount of consumption.

The determination OCR_0 involves putting coal and dry air (standard condition) in an airtight container and measuring the temporal change in oxygen concentration. This method was used to measure the relationship between the OCR and integrated oxygen consumption for bituminous coal from Australia (Coal A), as well as for sub-bituminous coal from Indonesia (Coal B). The results are shown in **Fig. 4**. As is evident from this figure, Coal B, sub-bituminous coal, has an OCR higher than that of the bituminous coal. The results also confirm that the OCR increases exponentially with increasing temperature. It should be noted that both types of coal show decreased reaction activity and



(a) Coal A (Bituminous Coal)

(b) Coal B (Sub-bituminous Coal)

Fig. 4 Oxygen consumption rate

a smaller *OCR* as the integrated value of oxygen consumption increases.

On the basis of the above, the *OCR* was measured for individual types of coals and was reflected in the simulation as a function, using Equation (3), of the integrated amount of oxygen consumption, temperature and oxygen concentration.

1.3 Evaporation, adsorption and desorption of moisture in coal

The evaporation, adsorption and desorption of moisture in coal are important in determining temperature behaviors inside a pile. This is because the moisture contained in coal evaporates, causing the latent heat of evaporation to cancel out the heat of low-temperature oxidation, which lowers the temperature inside the pile. The moisture contained in coal may be classified into two types: namely, (i) adsorption moisture existing in the pores of the coal, and (ii) adhering moisture caused by rainfall, the sprinkling of water, etc. Adsorption moisture is less prone to evaporate than is adhering moisture, and the amount of evaporation is determined from the relationship between the relative humidity and the amount of adsorption moisture.

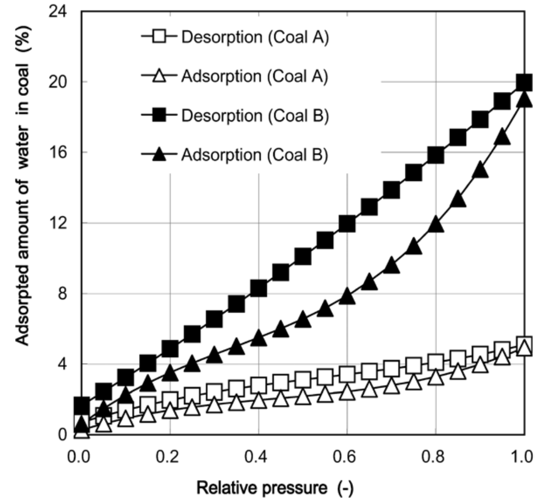


Fig. 5 Water adsorption/desorption isotherms

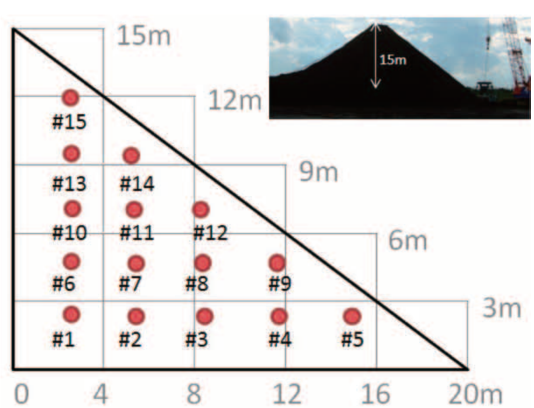
Fig. 5 shows the adsorption/desorption isotherms of water. These isotherms were obtained by putting coal and water vapor in a container with a variable pressure and by changing the pressure under isothermal conditions. These graphs represent values actually measured for bituminous coal (Coal A) and sub-bituminous coal (Coal B) under the condition of 40℃ (313 K). Under a given relative pressure, Coal B exhibits an increased amount of water adsorption, compared with Coal A. This is because the sub-bituminous coal has a larger specific surface area than bituminous coal; the coalification proceeds while changing the carbon structure and the chemical bonds of the coal. For a given humidity condition, only the moisture in the condition above the adsorption/desorption curve can evaporate, and this evaporation continues until this moisture is dissipated. When the relative pressure is constant, the moisture in the coal stays on or above the adsorption/desorption curve; meaning it does not decrease. In a condition where the heat generation is dominant, the temperature of the pile increases, decreasing the relative humidity. As a result, the evaporation continues, decreasing the moisture content of the coal. When the moisture in the coal is completely dried, no factor that can suppress heat generation exists any longer, and, as a result, the temperature in the pile rises faster.

On the basis of the above, the adsorption isotherms of water were measured for each type of coal. The evaporation behaviors of moisture in coal were more realistically modeled and simulated in this way.

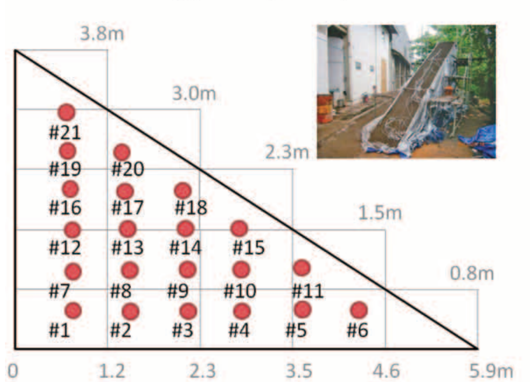
2. Measuring internal temperatures of test pile

In order to improve and verify the analysis accuracy, two test piles with different heights (15m and 4m) were prepared and their internal temperatures were measured (**Fig. 6**). The larger pile (15m pile) was cone-shaped and had a base angle of approximately 37°, a height of 15m and a bottom diameter of 40m. Measurements were performed at the 15 points shown in Fig. 6-(a) and the thermometers were spaced at intervals of 2.2m to 2.8m. The smaller pile (4m laboratory pile), on the other hand, was a partial cut-out of a cone-shaped pile and had the shape of a triangular prism with a height of 3.8m, width of 0.8m and bottom length of approximately 5.9m. Both of the triangle-shaped lateral faces were thermally insulated. Measurements were performed at the 21 points shown in Fig. 6-(b) at the center of the 0.8m width and the thermometers were spaced at intervals of approximately 0.5m to 0.7m.

The sub-bituminous coal, Coal B, was formed into the above two test piles and temperature measurements were conducted on each one for about a month. **Fig. 7** shows the results of these temperature measurements at typical points in the piles. As shown in Fig. 7(a), in the 15m large pile, the temperature rose remarkably at points #4, 5,

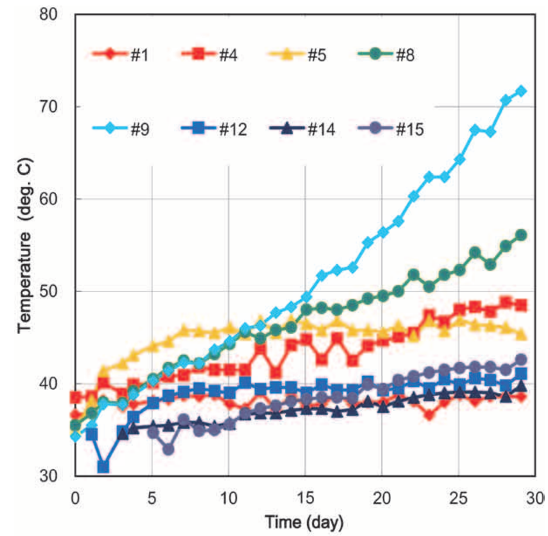


(a) 15m large scale pile

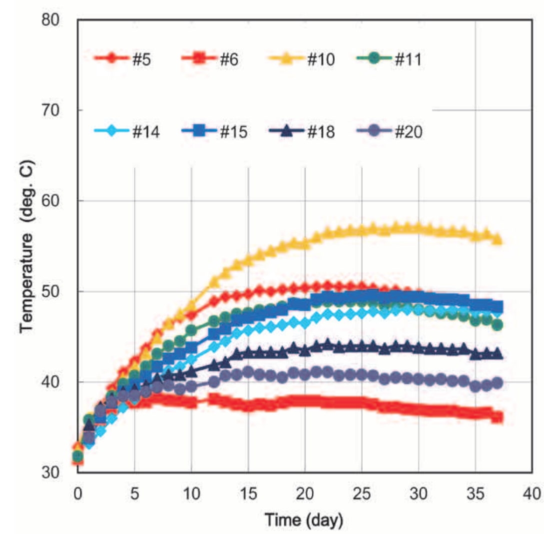


(b) 4m lab-scale test pile

Fig. 6 Measurement points of temperature inside pile



(a) 15m large scale pile for Coal B



(b) 4m lab-scale test pile for Coal B

Fig. 7 Measured results of temporal temperature change inside pile

8 and 9, each located at the foot of the pile. The heat is generated by oxidation, and oxygen must be supplied into the pile for it to occur. The remarkable temperature rise at the above points could thus be attributable to the fact that coal with a larger granule size tends to accumulate at the foot of the pile, as in the case of the 1.5m laboratory pile described in Section 1.1, making the breathability at the foot higher than at other points, enabling the supply of oxygen required for the heat generation. At the #9 position, in particular, the pile temperature rose at a rate of 1.2℃/d. This indicates that the temperature would reach 90℃ in 50 days. Once the pile temperature reaches 90℃, the temperature begins to rise abruptly. Thus ignition is presumed to occur at this location.

The 4m laboratory pile, shown in Fig. 7(b), also showed a remarkable temperature rise at points #5, #10, #11, #14 and #15, each at the foot of the pile as in the case of the larger pile. The maximum temperature reached in the case of the 4m laboratory pile, however, was lower than that reached by the larger pile. This is due to the greater amount of heat dissipated from the lateral faces of the triangular prism.

3. Prediction of heat generation based on pile simulation

The newly developed pile simulation was used to reproduce the heat generation behaviors in the two test piles (15m and 4m). **Table 1** shows the physical properties of the sub-bituminous coal from Indonesia (Coal B) used for the test. The physical properties of the bituminous coal from Australia (Coal A) are also shown for comparison. The O/C ratios−an index for spontaneous combustibility−are 0.05 and 0.14 respectively, indicating a higher spontaneous combustibility for Coal B. The major differences between Coal A and Coal B lie in the moisture content, specific heat, heat of oxidation reaction and activation energy. Coal B, containing a greater amount of moisture, is characterized by a higher specific heat and a smaller heat of oxidation reaction.

The initial conditions of the temperature and humidity for this simulation were taken from an average temperature and relative humidity corresponding to those of the atmosphere in Indonesia, where the temperature measurement experiments were carried out (**Table 2**). As for heat transfer behaviors inside a pile, the heat generation by oxidation reaction was estimated on the basis of thermal conduction, while the heat extraction from the pile by air flow was estimated on the basis of convection heat transfer.

Table 1 Coal properties

		Coal A, Australian bituminous coal	Coal B, Indonesian sub-bituminous coal
ρ	Density [kg/m^3]	1,150	1,100
C_p	Specific heat [kJ/kg/K]	1.46	2.30
λ_p	Heat conductivity [W/m/K]	0.25	0.25
D_p	Average diameter [mm]	5.0	5.0
ΔE	Activation energy [kJ/mol]	46	58
ΔH	Amount of heat generation [kJ/g-O_2]	10.73	6.91
ε	Void ratio [-]	0.19	0.19
W	Moisture [%]	11	27
O/C	O/C ratio	0.05	0.14

Table 2 Initial condition of coal pile and atmosphere

	Coal stock pile	Atmosphere
Initial temperature [deg. C]	30	30
Initial relative humidity [%]	88	70

The heat generated by the low-temperature oxidation reaction of coal is transferred inside the pile by thermal conduction in accordance with the effective thermal conductivity λ (W/m/K) defined by Equation (4). Here, ε represents the ratio of voids inside the pile, λ_p represents the thermal conductivity of coal and λ_g represents the thermal conductivity of air.

$$\lambda = (1-\varepsilon)\,\lambda_p + \varepsilon\lambda_g \quad \cdots\cdots (4)$$

The heat extraction from the pile, on the other hand, was assumed to be caused by natural convection (10W/m^2/K), since the velocity of the air flowing inside the pile is low. The dissipation of heat from the bottom of the pile to the ground was considered to take place by thermal conduction (10W/m/K). For the 4m laboratory pile, the amount of heat dissipated to the atmosphere from the triangular lateral face with insulation was assumed to be 0.5W/m^2/K. When measured, the amount of heat dissipated from the triangular lateral face of the 4m laboratory pile was, in fact, found to be approximately 0.5W/m^2/K to 0.6W/m^2/K.

The results of the simulation are shown in **Fig. 8** and **Fig. 9**. The objects of analysis were the 15m large piles of Coal A (Fig. 8, left) and Coal B (Fig. 8, right), and the 4m laboratory pile of Coal B (Fig. 9). The simulated data was compared with measured data, which proved that this pile simulation represents a good reproduction of the hot spots and the temporal change in temperature, regardless of the heights and shapes of the piles. In the case of the 15m large pile, the simulation has confirmed that the temperature rises significantly at the foot of the pile and as far as half-way up, in the same way as in the actually measured data, reaching approximately 70℃ to 75℃ after 30 days. The simulation has also been confirmed to be a good reproduction of the temperature that was reached, lowered by the dissipation from the triangular lateral face of a pile, e.g., one similar to the 4m laboratory pile, even if it has a different shape.

Comparing the 15m large piles of Coal A and Coal B, the simulation results show that Coal A reaches its maximum temperature of 50℃ after 30 days, while Coal B exhibits areas whose temperature exceeds 60℃ even after 15 days and reaches the proximity of 75℃ after 30 days. This is considered to be due to the high *OCR* of Coal B, sub-bituminous coal, which gives rise to a low-temperature oxidation reaction. In the case of Coal A, on the other hand, the temperature was found to rise in wider areas than in Coal B. This is considered to be attributable to the

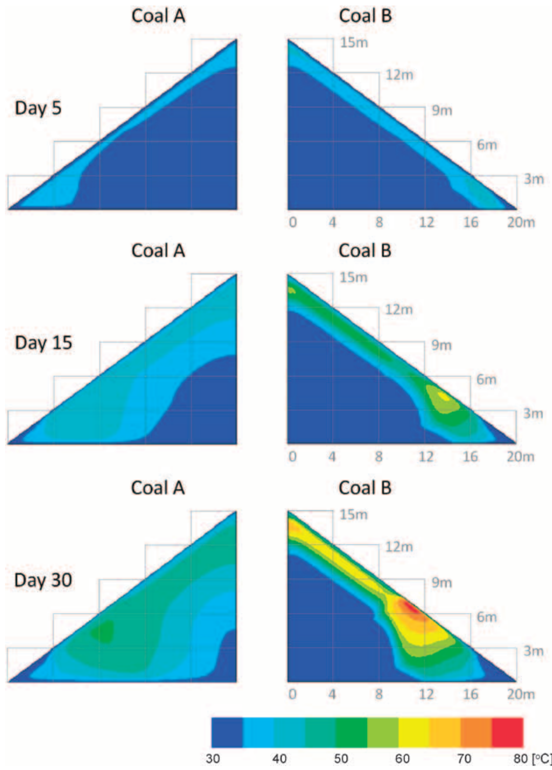


Fig. 8 Analysis results for temporal temperature change in 15m large piles of coal A and coal B

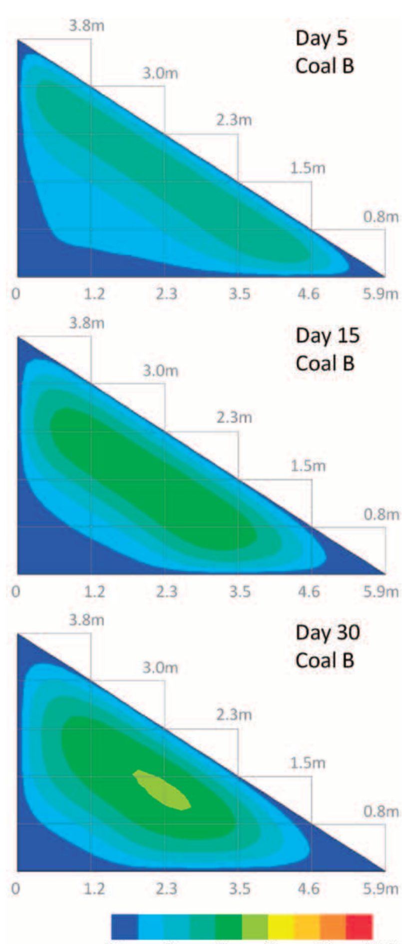


Fig. 9 Analysis results for temporal temperature change in 4m laboratory pile of Coal B

fact that Coal A has a lower specific heat and higher heat of oxidation reaction compared with coal B, which has a high moisture content (Table 1).

As described above, it was confirmed that the heat generation behavior during the storage of a new type of coal that has not been used before can be predicted without conducting trials on large-scale test piles. This can be achieved by laboratory tests to obtain data on the internal factors, including the low-temperature oxidation of the coal and the adsorption and desorption characteristics of water, and on the external factors, including the filling state of the pile and outer environment.

Conclusions

A heat generation simulation has been developed for piles of coal. In order to accurately reproduce the heat generation behaviors inside the piles, this simulation consists of unsteady analysis that links the heat transfer, flow and reaction, taking into consideration i) the flow behavior of air inside the pile, ii) the low-temperature oxidation behavior of coal inside the pile, and iii) the evaporation, adsorption and desorption behaviors of moisture in the coal. This simulation makes it possible to predict the heat generation behavior when storing a new type of coal that has not been used before.

In other words, the simulation allows predicting the spontaneous combustion of various new types of coal during storage by simply obtaining their physical properties through small-scale testing without conducting large-scale measurements involving several thousand tonnes of coal.

This technique is to be used to predict heat generation behavior during the storage of various new types of coals and to establish databases for appropriate delivery intervals.

References

1) Japan Oil, Gas and Metals National Corporation (JOGMEC). Research report on the development and advancement of coal from overseas. March. 2013.
2) H. Yamamoto et al. Journal of the Japan Institute of Energy. 2007, 86 (2), p.119-129.
3) H. Asaga. The thermal and nuclear power. 2006, 57 (6), p.426-430.
4) T. Muratani et al. Conference of coal science (38). 2001, p.279-282.
5) T. Ono et al. The thermal and nuclear power 33 (3). 1982, p.247-261.
6) T. Ono et al. The thermal and nuclear power 33 (4). 1982, p.379-386.