

A New Concept of Equivalent Oxidation Exposure-Time for Low Temperature Spontaneous Combustion of Coal

Kyuro SASAKI and Yuichi SUGAI

Department of Mining Engineering, Kyushu University
744 Motoooka, Nishi-ku, Fukuoka, 812-0395, Japan
e-mail: krsasaki@mine.kyushu-u.ac.jp

Abstract

This paper describes of a new concept of the equivalent oxidation exposure-time (EOE-Time) for spontaneous combustion of coal. The spontaneous combustions of coal have been studied, however, numerical models using with the Arrhenius equation applied to small amount of coal in low temperature range are not able to simulate actual heat and mass transfer phenomena. It still needs a numerical modeling to simulate heat generation rate in low temperature range. Some considerations of a kind of aging effect are required relating to oxidation capacity, cumulative oxidation amount, elapsed time in order to simulate heat generation rate of coal in low temperature range.

In this paper, the new analytical concept and calculation procedure of the EOE-Time to estimate the heat generation rate from coal have been proposed using a function of the EOE-Time, temperature and partial pressure of oxygen. The model follows some measurement results of coal heat generation rate for temperature and oxygen consumption. The present method has been applied successfully to simulate temperature rising processes from low temperature for coal seams and stock yards, which open-ends are exposed to the air using with a finite difference method by solving equations of heat transfer, oxygen diffusion and natural convection flow.

Furthermore, we have discussed the mechanism of spontaneous combustion of coal based on numerical simulation results showing time delay of oxygen diffusion compared with thermal diffusion in the coal seam and stock pile matrixes.

1. Introduction

Coal is a combustible material with a variety of oxidation scenarios from atmospheric temperature to ignition temperature. One of the most frequent and serious causes of coal fires is self heating or spontaneous combustion. Opening an underground coal seam to ventilated mine air such as long-wall gob areas and coal reserves in stockpiles have a risk of spontaneous combustion. Careful managements and handlings of coal stocks are always required to prevent fires. Furthermore, the spontaneous combustion also remains as a problem under transportations by marine ships and trains.

Generally, coal self heating has been explained based on a un-valance between heat transfer rate from a boundary surface to atmosphere and heat generated by oxidation in the stock. It is judged as a pre-stage of the spontaneous combustion when carbon monoxide exceeds a range of 100 to 200 ppm in the air around coal and its temperature becomes over a range 50 to 55°C. Thus, comprehensive studies on its mechanism and process of oxidation and temperature rising at low temperature range less than 50°C have been carried out for a long time. Measurement results on heat generating rate from crushed coal samples against constant temperature have been reported in order to evaluate a potential against spontaneous combustions. Kaji et al.(1987)

measured heat generating rate and gas adsorption rate of three kind of crushed coals under some constant temperature during 20 to 170°C using a calorimeter. They have presented a result that heat evolved per unit mole of oxygen at steady state is 314 to 377 kJ/mole.

According to observations in surface coal mines, spontaneous combustion of coal is appeared on coal surface as "hot spots". Usually, the hot spot have a root located at deeper regime from outside surface of a coal seam or a stock pile exposing to the air. When the hot spot is observed on the surface, its inside is smouldering due to low oxygen concentration. Heat generating rate from coal placed in high temperature range over 60 °C follows Arrhenius equation expressing a chemical reaction rate to accelerate a self heating. Brooks and Glasser (1986) have presented a simplified model of spontaneous combustion of coal stockpiles using with Arrhenius equation to estimate heat generating rate. They have considered a natural convection model as a reactant transport mechanism. However, when the Arrhenius equation is used for small coal lump, its calculation results of temperature do not show the return process to atmospheric temperature around it as shown in figure 1. A reason why the results can not applied to small amount of coal stock may be a kind of ageing effect that Nordon(1979) pointed out. He also has presented a model for self-heating reaction of coal and distanced two steady-state temperature conditions of less and over than 17°C.

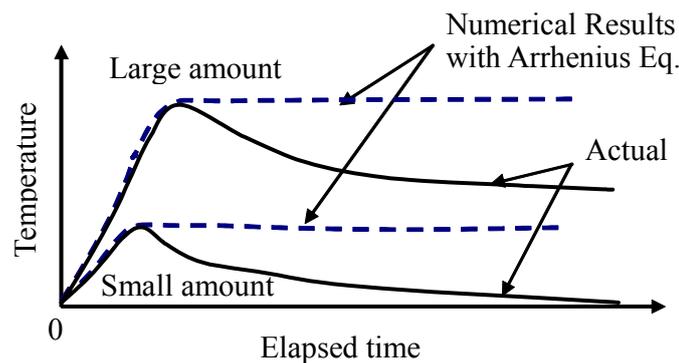


Figure 1 Difference of temperature change between a numerical simulation result by Arrhenius equation and actual process for small and large amounts of coal stock

In this study, a new scenario for spontaneous combustion in coal stock has been presented based on time difference between thermal diffusion and oxygen diffusion. Furthermore, new concepts of "Equivalent Oxidation Exposure-Time (EOE-Time)" has been presented. It like ageing time against its oxidation quantity in order to support the schematic mechanism presented. Numerical simulations matching with both thermal behaviors of large stock and small lump of coal have been carried out.

2. Mechanism of temperature rise in a large amount of coal stock

Coal exposing in the air is oxidized with adsorbing oxygen in low temperature range. It has different characteristics compared with oxidation expressed by the Arrhenius equation in high temperature range. The adsorption rate of oxygen is gradually decreasing with elapsed time when its temperature is kept as a constant, since the coal has an oxygen adsorption capacity.

As shown in figure 2, assuming coal stock is placed with exposing its outside to the air of θ_0 in temperature and C_0 in oxygen molar fraction concentration except its bottom surface, such as a coal stockpile. A kind of oxidation heat is generated at coal placed near the stock outside due to supplying oxygen from the air. The heat is not only lost to the atmosphere, but also it diffuses to inner direction of the stock. The outer regime of the stock is returned to atmospheric temperature, θ_0 , after enough time. On the other hand, the oxygen concentration of the inside stock is kept as a relatively low concentration, because oxygen is not provided through oxidation zone consuming oxygen at outer regime. The condition, that coal existing inside of the stock is preheated slowly without oxygen, generates high temperature spot of coal at its center regime.

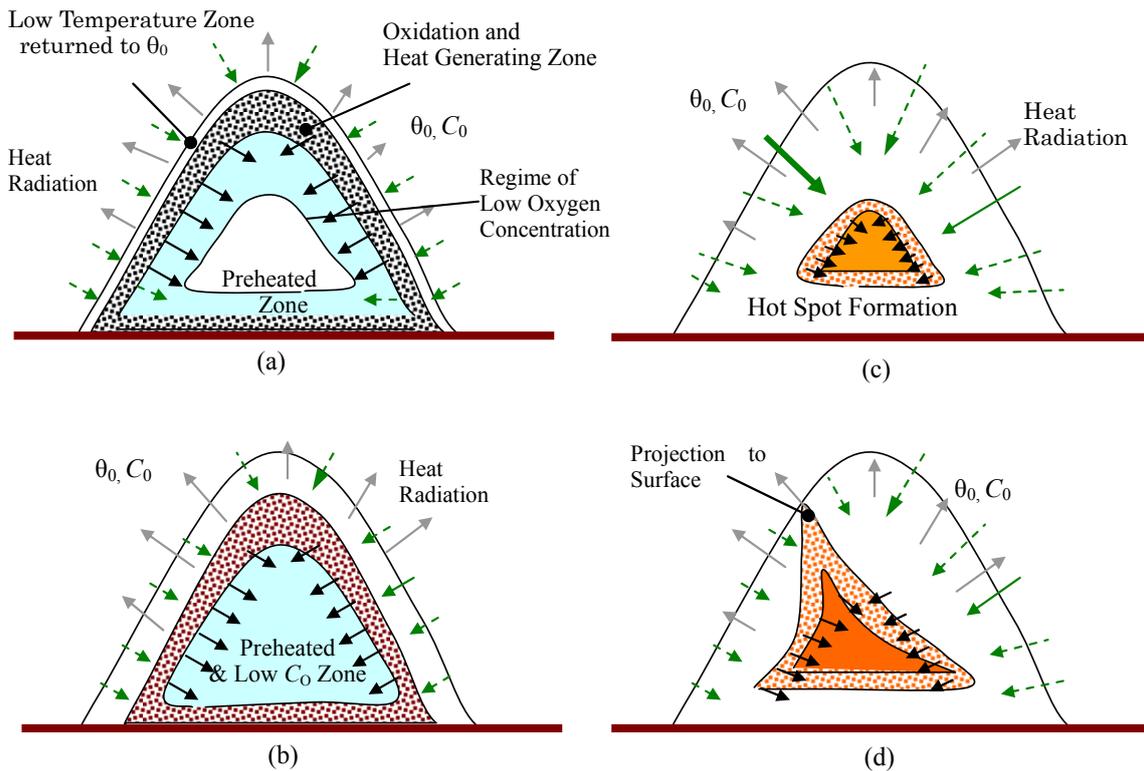


Figure 2 Schematic process showing spontaneous combustion of coal stock
 (a), (b), (c): Hot spot forming process with accumulating heat and shrinking regime of oxidation and preheating regime
 (d): Projection growth of hot spot toward to stock surface

The oxidation and heated regime is gradually moving from stock surface to center direction with shrinking its regime and rising temperature, and finally a hot spot is formed at the center regime(see figure 2 (a) to (c)). Oxygen diffuses into the center regime after formation of the hot spot. This time delay of oxygen diffusion towards the center regime after thermal preheating works effectively to make rising coal temperature exponentially in the center. Thus, more amount of coal stock, the more delay time it is of producing with enough preheating.

After formation of the hot spot in the center regime, it begins to project toward the outside through some paths with relatively high effective molecular diffusivity that oxygen is supplied from the outside of the stock. Finally, the hot spot appears on the outside surface of stock, then it is recognized as its spontaneous combustion.

3. Definition of EOE-time and heat generating rate of coal

3.1 Heat generating rate

In this paper, coal oxidation includes physical adsorption, chemical adsorption with oxygen reaction at low temperature range. Measurements of heat generating rate on early stage showing an exponential decrease have been reported by Kaji et al.(1987) and Miyakoshi et al.(1984). Based on their measurement results as shown in figure 2, heat generating rate per unit mass of coal at temperature $\theta(^{\circ}\text{C})$, q (W/g), can be expressed with a function of elapsed time after beginning of exposing to the air, τ (s),

$$q(\theta) = C \cdot A e^{-\beta\tau} \quad \dots(1)$$

where, A (kW/kg) is heat generating constant at $\tau=0$, and C is molar fraction of oxygen, and β (s⁻¹) is a decay power constant gives decreasing rate with the exponential function.

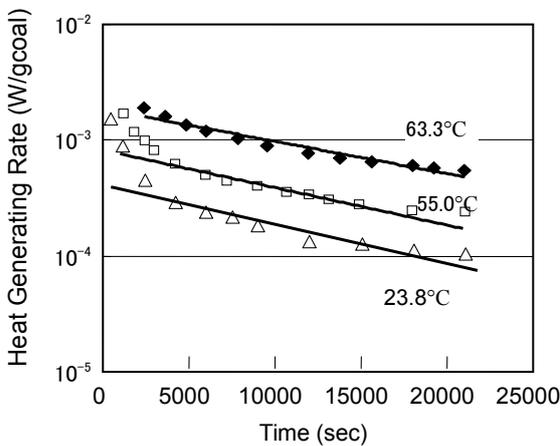


Figure 3 Heat generating rate of coal (Kaji et al., 1987) (Australia and bituminous coal)

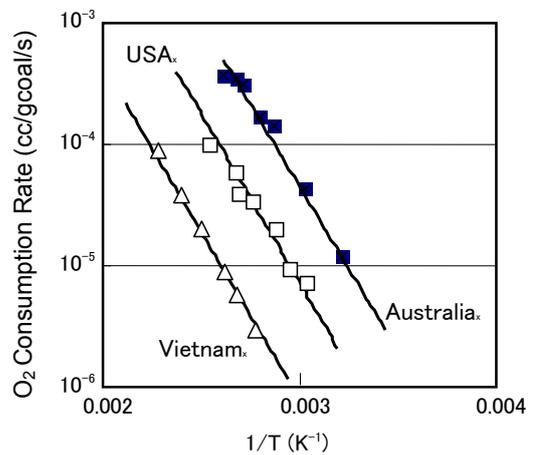


Figure 4 Oxygen consumption rate against absolute temperature (Kaji et al., 1987) (Australia bituminous coal, UAS sub-bituminous coal, and Vietnam smokeless coal)

3.2 Arrhenius Equation

Kaji et al. (1987) also presented measurement results of oxygen consumption rate against inverse of absolute temperature, T^{-1} (K⁻¹) (see figure 3). Their results show a relationship following the Arrhenius equation. Thus, the higher the coal temperature, the faster oxidation or adsorption rate will be generated. When the heat generating rate is proportional to oxygen consumption rate, A can be estimated with the equation,

$$A(\theta) = A_0 \cdot \exp\left(-\frac{E}{RT}\right) \quad \dots(2)$$

where, A_0 (kW/kg) is pre-exponential factor for heat generating rate, E (J/mole) is activation energy and R is gas constant, and T ($=273+\theta$)(K) is absolute temperature.

The heat generating rate is expressed with a function of θ , C , and τ . The equations (1) and (2) can be used to calculate q under a constant temperature, however, it is not applicable for the usual temperature change of coal against elapsed time. For a example, when coal lump is stored in an environment of $C = 0.1$ and $\theta=45^\circ\text{C}$, then moved to other one of $C=0.2$ and $\theta=70^\circ\text{C}$, it is not possible to make its elapsed time by adding the former and later times with different oxidation rates. A new concept of elapsed time considering aging degree of coal has been required to overcome the difficulty in applying the equation for constant temperature.

The cumulative generated heat of coal, Q'_m (J/g) during elapsed time 0 to t , is defined generally as,

$$Q'_m = \int_0^t q'(\theta', C', t') dt' \quad \dots(3)$$

where, actual heat generating rate, $q'(\theta', C', t')$, θ' and C' are changing with elapsed time t' . On the other hand, cumulative heat generated, Q_m , under constant θ and C , can be derived from Equations (1) and (2) during 0 to τ^* , as follows,

$$Q'_m = \int_0^{\tau^*} q(\theta, C, t') dt' = \frac{CA}{\beta} (1 - \exp(-\beta\tau^*)) = Q_m \quad \dots(4)$$

If the amounts of accumulated heat, Q'_m and Q_m , defined with Equations (3) and (4) are equal (see Figure 4), τ^* in Equation(4) expresses a kind of aging time of coal under the constant condition, $\theta=\theta'(t)$ and $C=C'(t)$, at actual elapsed time($t'=t$). In this paper, τ^* is called as EOE-time. It is calculated as the following equation,

$$\therefore \tau^* = -\frac{1}{\beta} \ln\left(1 - \frac{\beta Q'_m}{CA}\right) \quad \dots(5)$$

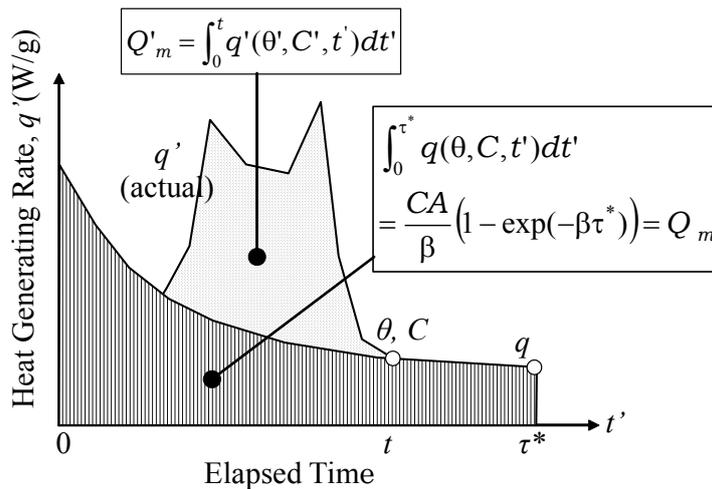


Figure 4 Schematic figure to define EOE-time

Based on the EOE-time, the actual heat generating rate of coal at $t'=t$ is obtained by substituting τ^* instead of τ into Equation (1). Assume the reaction heat of unit volume of oxygen is ΔH (J/cm³O₂), oxygen consumption rate, ν (cm³O₂/s·g), and accumulated consuming oxygen, V (cm³·O₂/g), of the coal are approximately given by next equations, respectively.

$$\nu = q / \Delta H \quad \dots(6)$$

$$V = \int_0^{\tau^*} \nu(\theta', C', t') dt' \quad \dots(7)$$

4. Numerical simulation results and discussion

4.1 Simulation model of a coal seam in underground mines

As shown in Figure 5, a simple one dimensional numerical model for a underground coal seam has been investigated to confirm a effectiveness of the EOE-time. Its faces are open to mine ventilated air with little ventilation pressure difference, $\Delta p = 10 \text{ mm H}_2\text{O} = 98 \text{ Pa}$. Thus, oxygen is provided by not only molecular diffusion but also permeable flow between two faces. The coal seam of $L= 5.0\text{m}$ in the length has effective diffusion coefficient $De=6.7 \times 10^{-6} \text{ m}^2/\text{s}$ and permeability $K_0= 50\text{md}$. Almost oxygen diffuses from both ends of the coal seam and adsorbed in the coal seam on the way of diffusion toward its center regime. Thermal and heat generating properties of the coal seam used in the simulations are listed in Tables 1 and 2. In this numerical simulation study, effects of moisture content of coal on heat generating rate and temperature rise are not considered. However, Sasaki et al.(1992)(one of authors) has presented its physical modeling and its effects.

The numerical simulation result on temperature distribution is shown in Figure 6. The regime of rising temperature and consumption oxygen is moving toward center with higher maximum temperature. On the other hand, outer regime temperature near the boundary surface to the air is becoming low, due to increasing EOE-time and heat loss to outside.

density of oxygen in the coal seam gradually diffuses to the center of the coal seam along with lapsed days. Moreover, there is the maximum value of the temperature distribution in the vicinity of both ends where the density receives the supply of oxygen easily high from the relation to such distribution of the density of oxygen to about the 90th. The position moves to the center part with the progress of the oxygen diffusion, and, in addition, the maximum temperature rises, too. This shows the mechanism to which the delay of the diffusion of the oxygen explains by paragraph 2 promotes the rise in heat of coal.

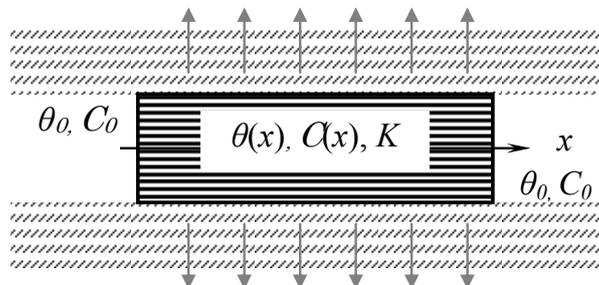


Figure 5. Numerical simulation model of a coal seam using one dimensional model

Table 1 Thermal properties of coal for numerical simulations shown in Figure 1

Thermal diffusivity	Density	Specific Heat
κ	ρ_0	C_p
$9.1 \times 10^{-8} \text{ m}^2/\text{s}$	1291 kg/m^3	$1.21 \text{ kJ/kg} \cdot ^\circ\text{C}$

Table 2 Heat generating properties of coal for numerical simulations shown in Figure 1

decay power constant	pre-exponential factor	activation energy
β	A_0	E
0.00329 s^{-1}	14.5 kW/kg	20 kJ/mol

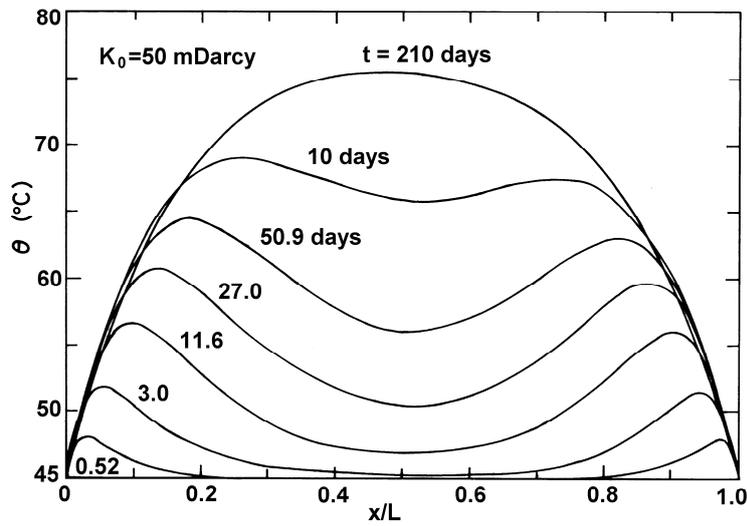
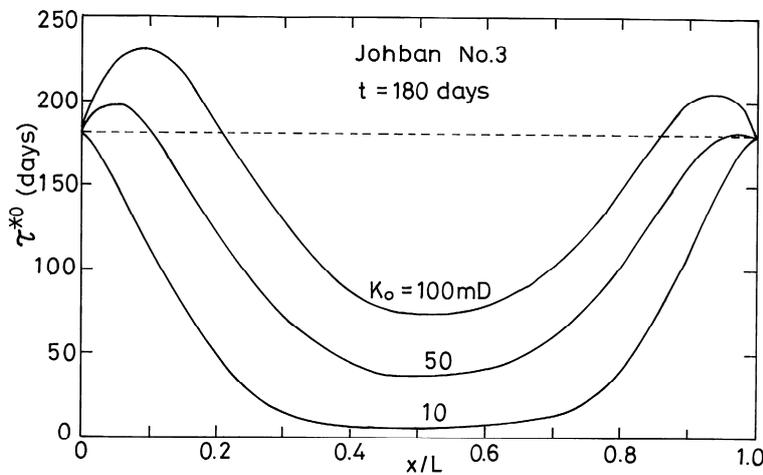
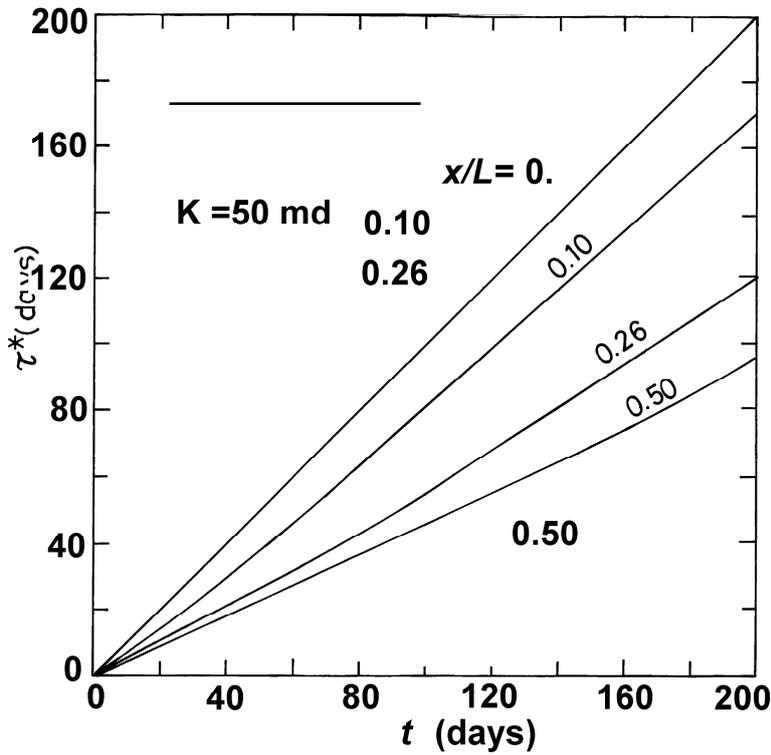


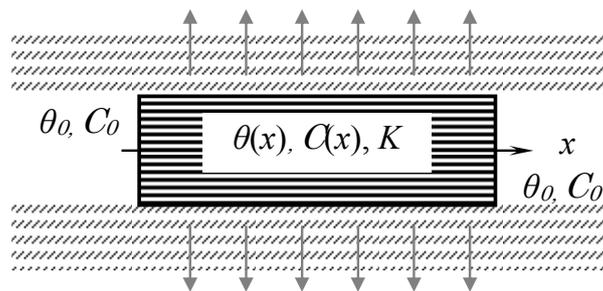
Figure 6. Transition of temperature distribution of coal seam



4.2 Numerical simulation result of coal stockpile

In the stock coal seam or the coal piling up seam, the air seam causes the natural convection along with the rise of an internal temperature, and it has a complex temperature rise mechanism that promotes the amount of the heat material movement of the oxygen supply and the heat movement, etc. further. The temperature rise assumption was examined according to two dimension numerical simulation model by whom the natural convection in the rectangular piling up seam shown in Figure 6 of the thermal storage process in consideration of the natural convection model corresponding to the humus-decay accumulative seam was considered. Half

the one side of the stock coal seam is made an analytical object, width from coordinate origin 0 is defined, and the height of L(m) and the direction of the perpendicular is defined as H(m). The elapsed time after it piles up is does ..streamline of the natural convection in the stock coal seam after 500 hours.. cloth and shows the temperature distribution in Figure 7 and Figure 8 respectively. It is understood to flow external air's flowing in from the side in the stock coal seam almost squarely, and toward the upper part of the seam. As for the temperature, it is understood that a low, high temperature region exists in the vicinity of the upper part of the center of the seam in the side part in which air from the outside flows in in the stock coal seam. Figure 7 shows the transition of the time of the maximum temperature in the coal piling up seam. Thermal diffusivity and the oxygen supply by the natural convection are shown.



Pattern diagrams of Figure 4 coal seam and tunnel

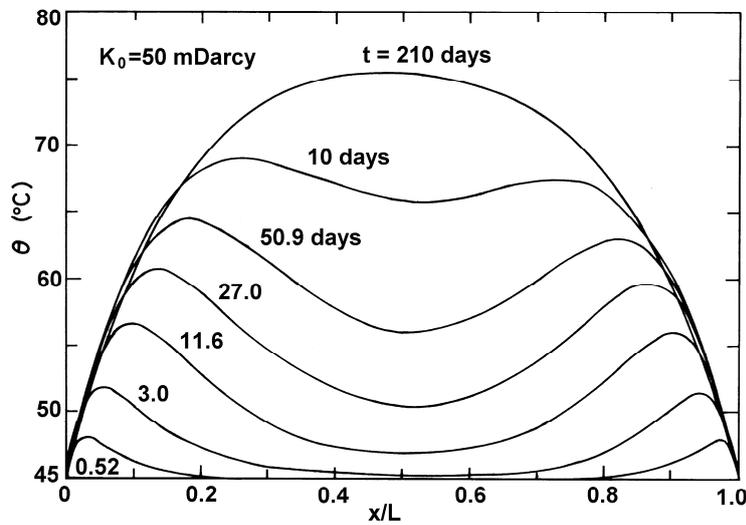


Figure 5 Transition of temperature distribution of coal seam (Sasaki et al.. 10)

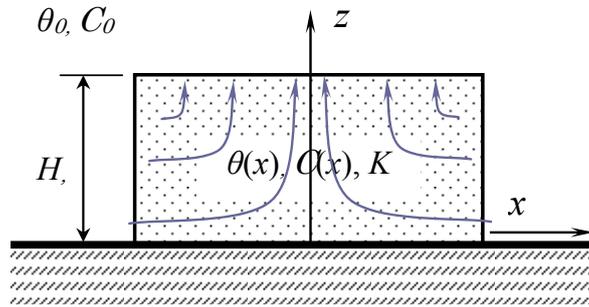


Figure 6 Pattern diagrams of coal seam and tunnel

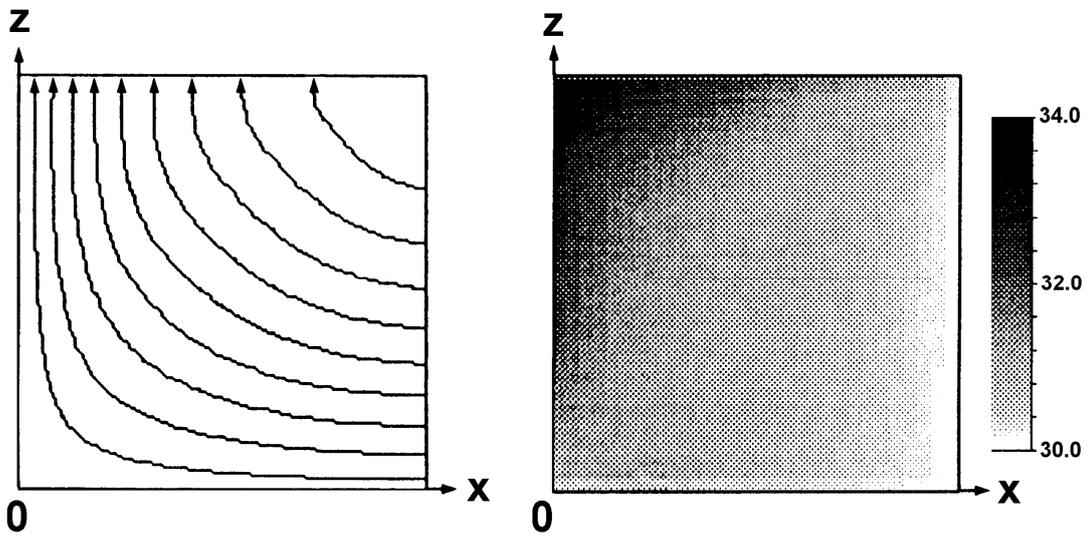


Figure 7 streamline of the natural convection according to generation of heat o coal.. cloth distribution ($L=5m, H=5m$) of the temperature.

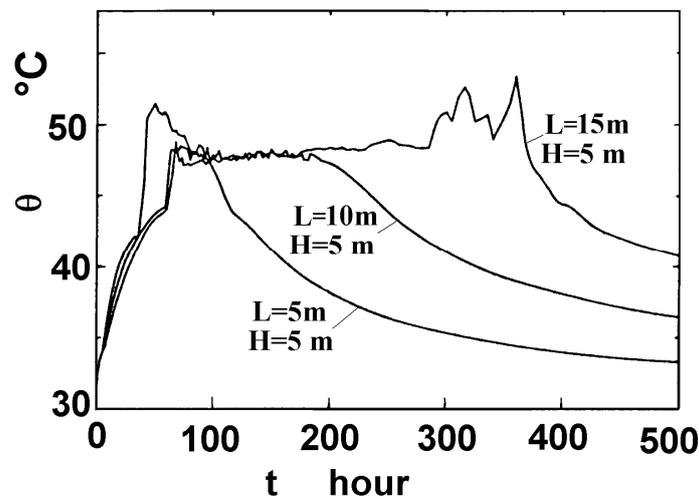


Figure 8 Transition at time of the maximum temperature in coal piling up seam

5. Summary

This paper describes of a new concept of the equivalent oxidation exposure-time for spontaneous combustion of coal. The spontaneous combustions of coal have been studied, however it still needs a numerical model to simulate heat generation rate in low temperature range. Heat generation by oxidation of coal in low temperature includes very complicate functions of temperature and time decay. Thus, numerical models using with the Arrhenius equation in considering coal temperature are not able to simulate actual heat and mass transfer phenomena. We have proposed the new analytical concept and calculation procedure of the equivalent exposure-time to estimate the heat generation rate from coal function of the equivalent exposure-time, temperature and partial pressure of oxygen. The model follows some measurement results of coal heat generation rate for temperature and oxygen consumption. The present method has been applied successfully to simulate temperature rising processes from low temperature for coal seams and stock yards, which open-ends are exposed to the air using with a finite difference method by solving equations of heat transfer, oxygen diffusion and natural convection flow.

Furthermore, we have discussed the mechanism of spontaneous combustion of coal based on numerical simulation results showing time delay of oxygen diffusion compared with thermal diffusion in the coal seam and stock pile matrixes.

The preheating advanced while reducing the area by the conduction of heat from the area in the vicinity of the edge side to which heat began to be generated previously as a reason why the temperature rises easily in the coal seam or the center part of the piling up seam, and the center part proposed the process to the ignition by the oxygen supply by diffusion afterwards.

Reference

- 1) Miyacoshi hiroshi, Toshiro Isobe, and Kazuo Otsuka: Relationship between oxygen adsorption and physico-chemical properties of coal, *IJournal of MMIJ*, Vol. 100 1161 and 1057-1062 (1984).
- 2) Sondr E. A. et al, & Ellman, R.C.: Report of Investigations No.7887, Bureau of Mines, Washington D. C.(1974) .
- 3) Italian Kishouge and Scocoroza Higuchi: *Journal of MMIJ*, Vol. 90 1033 and 175-180 (1974).
- 4) Toshiro Isobe, Kazuo Otsuka, and Miyacoshihiroshi: *Hokkaido mine society magazine* 24 4 and 16-22 (1968).
- 5) Kiyoshi Hashimoto: *16 Hokkaido mine society magazine* 16 1, 2, and (19-60).
- 6) Sasaki, Miyakoshi, and Otsuka: Rally lecture summary collection and 265-267 in *Nippon Mining Company, Limited association spring* in 1986 fiscal year.
- 7) Chamberlain, E. A. C. : *Colliery Guardian*, 233[3], 79~82 (1974)
- 8) Peters, W.:*Gluckauf Jg. 101 Heft 26,1526~1531* (1965)
- 9) Kaji, R. et al.: *Fuel*, Vol.66 Feb., 154~157 (1987)
- 10) Sasaki, Miyakoshi, and Otsuka: *Journal of MMIJ* Vol.103-1197, 771-775.
- 11) K. Sasaki et al., Water vapor adsorption of coal and numerical simulation related its effects

on spontaneous combustion in a low temperature range, Journal of MMIJ, Vol.108-6, pp.479-486, 1992(in Japanese)

Nordon, P., "A Model for the Self-Heating Reaction of Coal and Char", Fuel, 58, 456-464, 1979.

Brooks, K. and Glasser, D., "A Simplified Model of Spontaneous Combustion in Coal Stockpiles", Fuel, 65, 1035-1041, 1986.

Computational Fluid Dynamics Modeling of Spontaneous Heating in Longwall Gob Areas

In order to provide insights for the optimization of ventilation systems for U.S. underground coal mines facing both methane control and spontaneous combustion issues, a computational fluid dynamics (CFD) study was conducted to model the potential for spontaneous heating in longwall gob areas.

Yuan-L, Smith-AC , Computational Fluid Dynamics Modeling of Spontaneous Heating in Longwall Gob Areas, 2007 SME Annual Meeting and Exhibit, February 25-28, Denver, Colorado, preprint 07-101. Littleton, CO: Society for Mining, Metallurgy, and Exploration, Inc., 2007 Feb; :1-7, NIOSHTIC-2 No. 20031657

