

COUPLED MULTI-STAGE OXIDATION AND THERMODYNAMIC PROCESS IN COAL-BEARING STRATA UNDER SPONTANEOUS COMBUSTION CONDITION

Yanming Wang^{*,**,§}, Guoqing Shi^{*,**} and Zhixiong Guo^{**}

^{*}School of Safety Engineering, China University of Mining and Technology, Xuzhou, China.

^{**}Dept. of Mechanical and Aerospace Engineering, Rutgers, The State University of New Jersey, Piscataway, USA.

[§]Correspondence author. Email: cumtwangym@163.com

ABSTRACT Spontaneous combustion of subsurface coal is an extremely complicated chemical and physical changing process. In order to quantitatively understand the development of underground coal fires, the heat transfer processes coupled with multi-stage oxidation of coal are investigated by experimental and numerical methods in this paper. With controlled temperature and under lean oxygen conditions, the chemical thermodynamic parameters of coal oxidation on different stages are measured utilizing the simultaneous thermal analysis. Employing the composite boundary conditions, a heat transfer model with finite reactions for porous coal and rock is developed to simulate the thermal dynamical process in coal-bearing strata. The temperature fields of coal and roof strata at different times are estimated based on single- and two-stage oxidation kinetic models, respectively. The effects of the oxidation kinetic properties due to coal metamorphism on propagation of coal fires are examined and compared. It demonstrates that there exists a significant step change during the thermal process of coal fire caused by multi-stage oxidation, and the coal rank of occurrence directly determines the spontaneous combustion period of underground coal fire.

KEYWORDS Coal fire, Oxidation model, Thermodynamic process, Heat transfer.

INTRODUCTION

Uncontrolled coal fires are a serious problem in many coal-production countries and it has become an environmental and economic problem of international magnitude [Stracher 2009]. Although written accounts of coal fires date back to at least the time of Alexander the Great, the worldwide spread of coal fires has increased dramatically since the industrial revolution [Whitehouse and Mulyana 2004]. Currently, thousands of coal fires are burning - some for centuries and many uncontrollably, with flames up to 20 m and temperatures exceeding 1000 °C - from eastern Asia and northern China into the coal basins of Russia, Europe, Africa, north and south America, and Australia [Stracher and Taylor 2004].

Coal fire is a global catastrophe. Some of its prime impacts are: 1) Emission of many toxic gases, such as carbon monoxide (CO), carbon dioxide (CO₂), sulfur oxides (SO_x), methane (CH₄), and nitrogen oxides (NO_x). Among these noxious gases, CO₂ and CH₄ contribute to global warming. 2) Geomorphic effects, including land subsidence, surface cracks, faults, and other geologic structures. 3) Temperature increment of surrounding areas, desiccation of forests, lowering of water quality. 4)

Wasteful consumption of a potentially valuable resource. 5) Increment of production cost due to fire extinguishing and difficulties in mining operations.

Spontaneous combustion of coal is the main cause of coal fires. In recent years, work in the field of coal spontaneous combustion was mainly focused on heat release, gas products, the functional groups and free radical changes of the spontaneous combustion of coal [Jones et al. 2003, Ozbas et al. 2003, Beamish and Blazak 2005, Yip et al. 2011]. Meanwhile, high temperature combustion and the pyrolysis characteristics of coal are the research hotspots in the energy chemical industry; and the oxygen-enriched combustion characteristics of coal such as initial reaction temperature, ignition temperature, combustion reaction rate and their relationships with the oxygen concentration have been studied intensively [Porada 2004, Biswas et al. 2006, Gil et al. 2010, Taraba et al. 2011].

However, the susceptibility to spontaneous combustion depends not only upon the material but also its physical state as well as the psychometric condition and migration paths of the leakage airflows. The development of self-heating requires a large surface area of crushed material, as well as a slow migration of air through that material. Hence, spontaneous combustion of coal underground takes place mainly in the strata areas, caved zones, and crushed pillar edges, and occurs through a complex system of thermal, hydraulic, chemical, and mechanical processes. Many efforts have been undertaken to understand the dynamic processes involved in coal combustion, which are of physical and chemical nature [Rosema et al. 2001, Huang et al. 2001, Wolf and Bruining 2007, Wessling et al. 2008]. Despite their potential for environmental impacts, coal fires are not accurately quantified in terms of temperature predictions. Because the multi-stage process of coal oxidation was not considered in prior works, it could cause the ignition temperature occurring prematurely and that simulation results might not be consistent with in-situ measurements.

In order to understand the thermal risk caused by coal fires in subsurface caved strata accurately, this study investigates the process of oxygen chemisorptions on coal in different temperatures using TG-DSC apparatus firstly. Heat evolution during oxygen chemisorptions and kinetic parameters are evaluated under different temperature levels. Then, the coupled heat transfer model with finite reactions for porous coal and rock media is established. Combined with the oxidation kinetic parameters, the thermal dynamical process in coal-bearing strata is simulated. The temperature fields of coal fire at different times are estimated. And also, the effects of oxidation kinetic feature due to coal metamorphism on propagation characteristics of coal fires are examined.

EXPERIMENT

Mechanism of Coal Oxidation The phenomenon of spontaneous combustion has been recognized since at least the seventeenth century. An early theory postulated that oxidation of pyretic material in coal provided a center of enhanced activity. This can play a part in the spontaneous combustion of timber or organic waste material underground but is unlikely to contribute significantly to the self-heating of coal or other minerals. Current researches on the initiation of self-heating showed that coal spontaneous combustion arose from the combined effects of coal and oxygen, including physical adsorption, chemical adsorption and chemical reaction.

Oxidation of coal takes place after four stages [Pone et al. 2007]: 1) physical adsorption of oxygen, resulting in temperature increase; 2) chemical adsorption (over 50 °C), producing oxygenated hydrocarbons or peroxy-complexes; 3) decomposition of oxygenated hydrocarbons when self-heating temperature is reached (over 70 °C), with concurrent oxidation of unaltered coal matter; and 4) spontaneous combustion may then occur if all the above processes result in a temperature higher than 150 °C, typically referred to as the ignition threshold. This ignition threshold seems to depend on coal rank, with values around 130 °C for the lowest rank, and probably even lower values for peat. In this respect, the 60-80 °C range is regarded as critical [Misz et al. 2007].

Oxidation Kinetic Properties Of the above processes, the coal and oxygen react and release gaseous products such as carbon monoxide (CO), carbon dioxide (CO₂), and water vapour (H₂O). Coal spontaneous combustion can be regarded as an exothermic reaction of coal and oxygen at low concentration as follows:



The reaction rate is predominated by the concentration of oxygen in the fire front area, which is mainly supplied by air convective transport through the fractures of overlying rocks. According to the Arrhenius equation, coal reaction rate at different temperatures can be expressed by the following equation.

$$\omega = -C_c A e^{-E/RT} \quad (2)$$

Where C_c is the coal concentrations, E and A are the activation energy and the frequency factor depending on the coal rank and measurement method, R is the gas constant and $R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$, and T is the absolute temperature.

TG-DSC measurement enables evaluation of the heat evolved during the oxygen chemisorptions process by relating the heat to the mass increase in the same temperature interval [Jones et al. 1998]. In this study, simultaneous thermal analysis was carried out in a SDT-Q600 instrument which was used in the thermo gravimetric analyzer (TGA) and differential scanning calorimetry (DSC) configuration. A brief experimental procedure is as follows [Li et al. 2014]:

- 1) Before the experiments, weight, temperature and sensitivity calibrations of the instrument were performed using calibration sets provided by SDT.
- 2) About 8mg of a vacuum-dried coal sample of 180-380 μm was placed in the reference crucible made of α -Al₂O₃. Here, three kinds of typical coal were involved in the present measurements as case studies where samples A, B and C were long flame, bituminous and anthracite coal, respectively.
- 3) The sample was firstly heated to 110 $^{\circ}\text{C}$ in pure N₂ with a flow rate of 70mL/min, held for 330 minutes to further drive off the moisture and followed by cooling down to 30 $^{\circ}\text{C}$.
- 4) And then, hybrid gas of ultra high purity O₂ and N₂ with 15% of O₂ was switched to air with a flow rate of 100mL/min. Beginning at 30 $^{\circ}\text{C}$, the sample was heated to 500 $^{\circ}\text{C}$ with a constant heating rate of 1 $^{\circ}\text{C}$ per minute.

The profiles of weight (denoted as TG curve), heat evolution rate (denoted as DSC curve) of coal C varying with temperature are illustrated in Fig. 1 to show typical results from the TGA-DSC test on non-isothermal oxidation of a coal sample.

Based on TG and DSC data, the kinetic parameters (critical temperature T_c , activation energy E , frequency factor A) of multi-stage coal oxidation could be determined by nonlinear regression from TG with presumption of the Fn kinetic model [Zarrouk and O'Sullivan 2006]. For coal sample C, the activation energy below and above the critical temperature (102 $^{\circ}\text{C}$) are 86.2 and 38.1kJ/mol respectively while relevant values of frequency factor are 310 and 1.26×10^6 Hz.

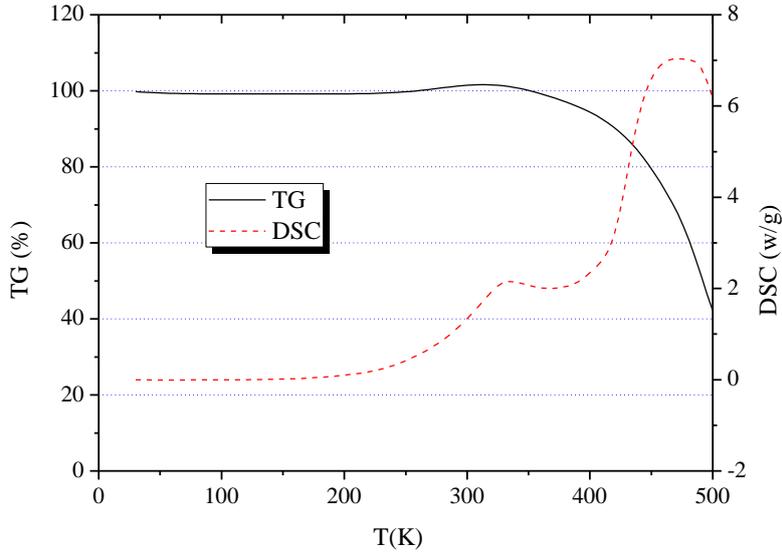


Figure 1. Profiles from TG-DSC measurement of a typical coal.

MODELING

Mathematical Model The mathematical model for temperature distribution in the coal and rock strata is developed along with specific boundary conditions and an initial conditions. The following equations apply to the coupled diffusion-reaction-heat transfer phenomenon in strata.

The energy transport equation is formulated under the assumption of thermal equilibrium between the solid matrix and gas. Thermal energy released by coal spontaneous combustion is transported through three mechanisms: heat conduction through surrounding rocks, convection through gas transport in the rocks, and thermal radiation due to high combustion temperatures. Because coal oxidation is an exothermic process, the link between heat production and oxygen consumption must be considered. Therefore, the mathematical model should contain energy conservation equations below.

$$C_c C_p \frac{\partial T}{\partial t} + \nabla \cdot q_h - Q = 0 \quad (3)$$

Where t is the time, C_p is the specific heat capacity of coal, T is the thermodynamic temperature, q_h is total heat flux, and Q is the source term which describes the heat release capacity of coal. The energy source term is directly proportional to calorific value ΔH through coal consumption rate ω . The heat flux consists of three parts: heat conduction, convection, and radiation, which can be formulated as:

$$q_h = q_{cd} + q_{cv} + q_r \quad (4)$$

The conduction heat flux is given according to Fourier's law:

$$q_{cd} = -\lambda \nabla T \quad (5)$$

The convection heat flux q_{cv} through gas transport in the strata can be formulated as:

$$q_{cv} = C_f C_d^f \nabla T \quad (6)$$

Where λ is the thermal conductivity of coal or bulk in the strata, C_f is the gas concentration, Cd^f is the convective heat transfer coefficient and v is the seepage velocity in the strata which is determined by the strata permeability and oxygen distribution. In this study, the seepage velocity is related to the difference value of oxygen concentration from contiguous areas in the strata.

The radiation heat flux q_r is determined by radiative transfer equation (RTE). Many methods have been established and applied to solve RTE equation successfully [Tan et al. 2006, 2009]. Here, considering the efficiency of coupled computation, a relatively simple 4-flux model is adopted in the 2-D simulation.

Simulation Setup The two-dimensional simulation domain of the subsurface coal fire propagation is given in Fig. 2. The length of the strata in the model is 200m, and the whole height from the surface to the floor is 100m. The coal seam (zone A) is embedded 50m deep and is 2m thick. The finite difference method with implicit numerical scheme is implemented in the heat transfer simulation. A regular grid is carried on the simulating domain in which the general interval is set to 1m and more refined grids are applied to the coal seam zone.

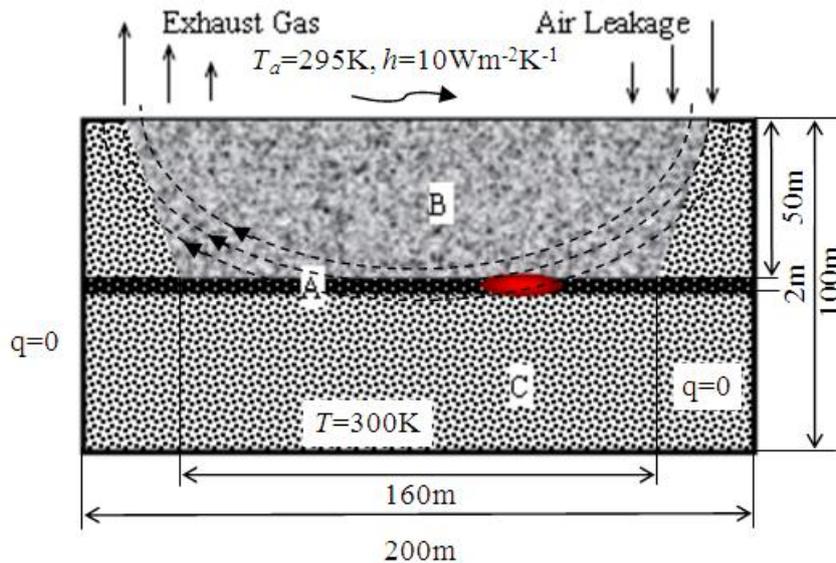


Figure 2. Sketch of the strata used in the present simulation. (Zone A: coal seam, Zone B: caved roof, Zone C: rock floor)

There exists two main fracture channels at the edges of top subsidence caused by mining: the airflows into the crush rock from one side and the oxygen is supplied into coal seam by air seepage. The oxygen concentration has great influence on the thermodynamic properties of the oxidation and combustion of coal. Generally, the oxygen concentration decreases along the whole leakage channel when the airflows through the coal seam because of adsorption function of low-temperature coal oxidation. Therefore, a liner decreasing distribution of oxygen concentration is implemented which means that the highest concentration in the coal seam exists near the air entrance fracture and the lowest value exists near the gas exhaust channel. And then, the seepage velocity is assumed as a function of difference value of oxygen concentration.

The main physical parameters employed in the heat transfer modeling are presented as followings: the specific heat capacity of rock is assumed as $4 \times 10^5 \text{Jm}^{-3}\text{K}^{-1}$, the thermal conductivity of coal and bulk are set to 0.2 and $2.0 \text{Wm}^{-1}\text{K}^{-1}$ respectively, the convective heat transfer coefficient between

coal and bulk is assumed as $1000\text{Jkg}^{-1}\text{K}^{-1}$, the densities of coal and bulk are 1800 and 2300kgm^{-3} respectively, and the porosity of these two materials are 0.5 and 0.2 respectively.

The boundary and initial conditions are defined below. At the top surface, Robin boundary condition is applied for the surface considering convective heat transfer between rock and atmosphere. Since the iteration step of simulation in this case study is usually set to 10 days during the coal self-heating development of several years, radiative heat transfer through the surface from solar or the earth day and nights is ignored. Here, the ambient temperature and convection coefficient are assumed 295K and $10\text{Wm}^{-2}\text{K}^{-1}$. Considering the symmetrical boundary on both sides of the simulation domain, Neumann conditions are implemented and the diffusive flux of thermal energy is assumed zero. Since the deep layer below the domain could be regarded as invariable temperature stratum, Dirichlet condition is applied for the bottom boundary in which the temperature is assumed 300K invariably. An initial temperature of 300K is assumed for all the zones in the strata.

RESULTS AND DISCUSSION

Thermal Dynamic Process First, the process of subsurface coal fire propagation is evaluated numerically. The two-stage oxidation kinetic parameters of coal sample C are used in the calculation as an example. Fig. 3 presents the simulated temperature distributions of the coal fire propagation at four consecutive time stages.

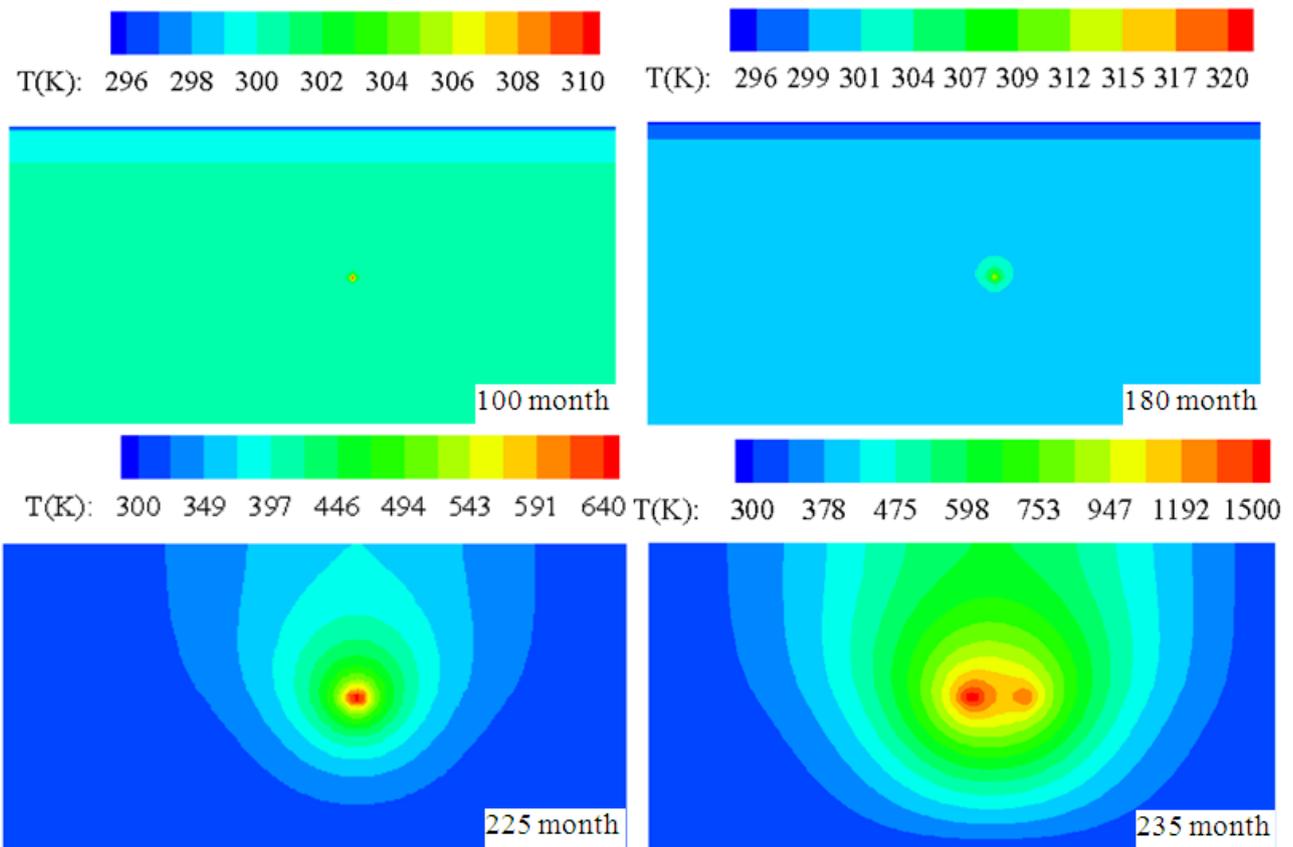


Figure 3. Temperature distribution due to the underground coal fire at different times.

The resulting temperature distributions show that the original temperatures in the coal and rock remain rather constant even after 15 years. It means that the heat diffusion process and temperature rise in the coal is very slow in this early period. When the temperature in oxidation core zone increases to over a critical value, a step change occurs in the thermal dynamic process in coal seam

and the heating rate shows a geometric growth from time at 225 months to 235 months. The combustion center first appears as a point-wise geometry and thereafter becomes elongated and then the elongated shape breaks down into two combustion centers that move in the up- and downward directions through the seam. The maximum temperature reaches to 1545K after 235 months by the upward moving fire, whereas the downward moving one exhibits a lower value.

Fig. 4 shows the temperature distributions for the locations below and above the coal seam as well as just at the surface at two different times. The temperature develops slowly above the ambient temperature. The distributions for both the below and above the coal seam locations are similar. Hence, the heat is not only released towards the surface, but also down into the rock. Meanwhile, the temperature below the seam is slightly smaller than that above the seam because of the different constraint conditions at the top and bottom boundaries. The pink vertical line coincides with the highest temperature and indicates the position of combustion core. Comparing the results at the two different times, it demonstrates that the fire front is moving slowly after the coal self ignition (roughly around 5m during 10 months). The figure also shows that the horizontal positions with the highest temperature for above the seam and at the surface are inconsistent during the development of coal fire.

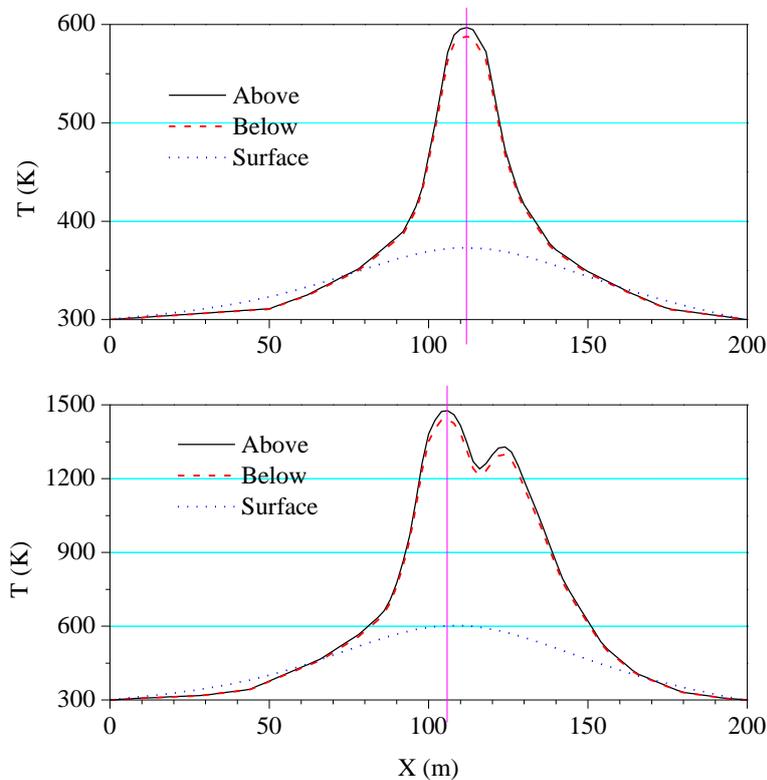


Figure 4. Temperature distributions above, below the coal seam and at the surface. (Top: simulating time at 225 month, Bottom: at 235 month)

Effect of Oxidation Model A more significant explanation for this problem is to trace the dynamic development of the maximum combustion temperature during the coal fire propagation. As illustrated in the curve of the multi-stage model in Fig. 5, a liner and extremely slow increment of maximum temperature in the strata is obtained up to over 180 months of coal oxidation. When the oxidation temperature increases beyond a threshold value, the chemical reaction process of coal is changing from the first-stage to the second-stage and the maximum temperature increasing rate proceeds much faster. It is seen that the oxidation temperature is over the critical value after about

210 months and then the coal seam starts to self ignite (above 350°C) about 15 months later. Finally, the temperature rises from ignition to more than 1500K in just 10 months.

In order to investigate the accuracy of the present two-stage model, a single-stage oxidation model is employed for comparison, in which the kinetic parameters adopt the second-stage values. Comparing the two curves in Fig. 5, it shows that the coal fire process (less than 2 years) simulated by the single-stage oxidation model is much shorter than that using the multi-stage model. Many reports in the literature have been documented that the history of coal fire is more than ten years after mining generally. For example, numerical modeling was developed to the analysis of bituminous coal seam burning [Klika et al. 2011] and the simulation showed that coal seam began burning after 100 years. And also, in-situ measurements which determined the combustion zones in abandoned underground coal mines [Kim 2004] reported that the combustion activity increased during a long-term period. Therefore, these evidences indicate that the multi-stage oxidation model coincides with the observation and simulation results better.

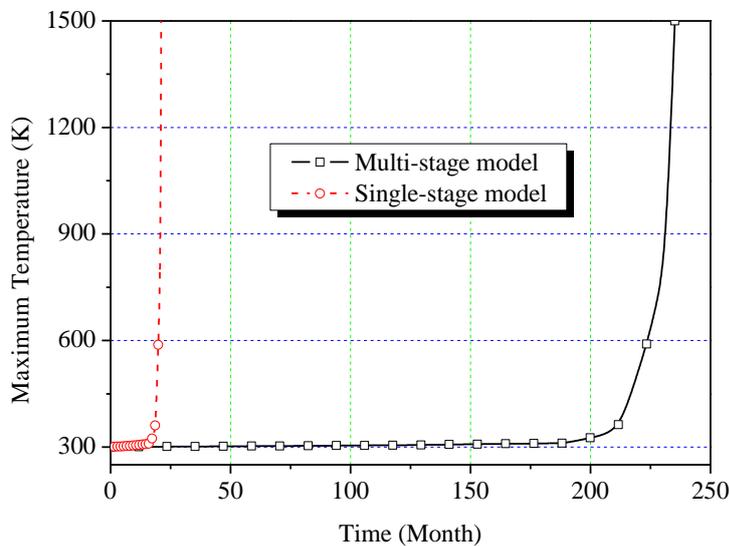


Figure 5. Dynamic development of the maximum combustion temperature.

To comprehensively understand the differences between oxidation models, the temperature distributions along the centerline of coal seam at different fire developing times are illustrated in Fig. 6. The time of coal temperature reaching self ignition by using the two-stage model is more than ten times than that by using the single-stage model. Using the single-stage model it takes only 2 month for the coal temperature close to 1500K after coal combustion, while it costs nearly 1 year based on the two-stage model. Meanwhile, the coal fire zone of the single-stage model is much wider than that predicted by the two-stage model. The pink dash dot line in Fig. 6 indicates the development of the fire front. It is seen that the fire propagation rate simulated by the one-step model is much faster than that by using the two-step model. Since previous surveys [Ide et al. 2011] by magnetometer measurements have reported that the combustion front of underground coal fire moved very slowly (advanced around 1m in a 5 months period), the multi-stage oxidation model is more adequate for predicting the long history of coal fire propagation.

Effect of Coal Rank In order to study the effects of coal metamorphism which measured of coal rank on the thermal dynamic process of coal fire, three kinds of typical coal mentioned above are involved in the present work. Here, sample A has the lowest rank while C has the highest. Kinetic parameters of these three samples determined by using simultaneous thermal analysis including the

averaged E , A and heat release Q below or above the critical temperature of certain coal are obtained and listed in table 1.

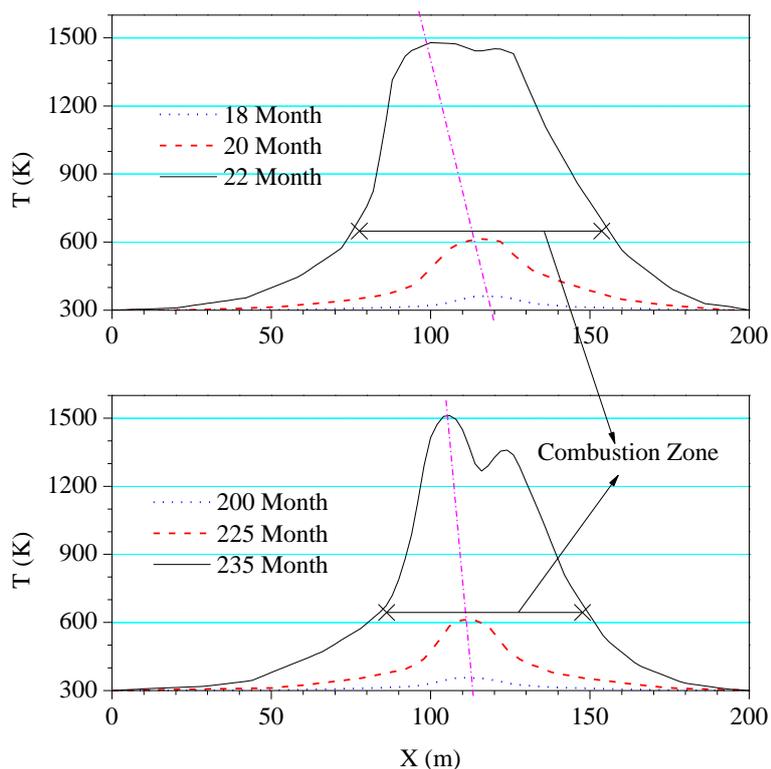


Figure 6. Temperature profiles at different simulation times along the centerline of coal seam. (Top: single-stage model, bottom: two-stage model)

Table 1
Oxidation kinetic parameters of the coal samples.

Sample	Carbon Content (%)	Critical Temperature (°C)	Stage	E (kJ/mol)	A (Hz)	Q (kJ/g)
A(long flame)	49.93	63	Below	31.9	120	2.2
			Above	63.3	13600	27.7
B(bitumite)	67.21	88	Below	34.2	170	2.3
			Above	77.5	340000	29.4
C(anthracite)	85.35	102	Below	38.1	310	3.1
			Above	86.2	1260000	32.5

It is remarkable that the data at the low-temperature below the critical temperature are very different than those in the high-temperature range above the critical temperature. In fact, in the low-temperature range the activation energy E and frequency factor A increase with temperature, while above the so-called threshold temperature a more stable reaction takes place. That means that the oxidation rate is very slow in the low-temperature range and changes very fast when the temperature of coal is across the threshold temperature.

Using the two-stage reaction model, Fig. 7 shows the time development of maximum temperature for the three coal samples listed in Table 1. It is seen that the heat rate of the low rank coal seam is rapid. Meanwhile the maximum combustion temperature of the high rank coal seam could achieve the higher level. These results are due to the difference of oxidation properties of different rank

coals. From Table 1, it is clear that the activation energy of the lowest rank sample is lower than those of the other two samples. It means that the oxidation process could occur earlier and more easily. On the other side, the highest rank sample has the largest heat release so that the spontaneous combustion of the coal fire could realize the highest maximum temperature.

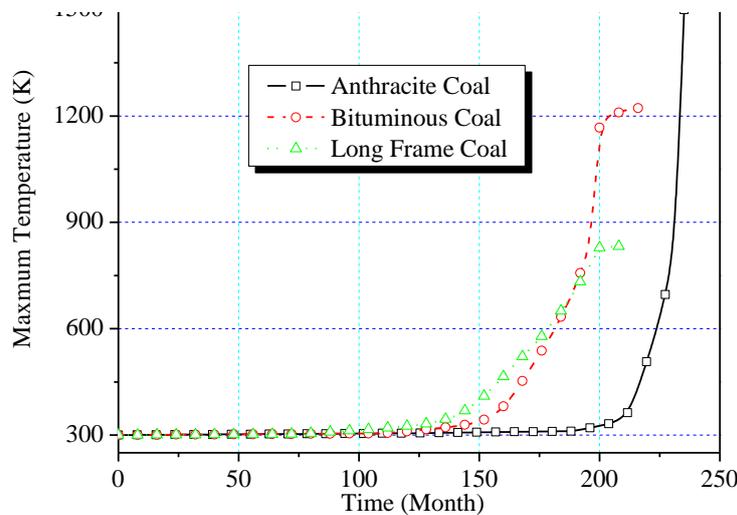


Figure 7. Development of the maximum temperature for different rank coals.

CONCLUSIONS

The effects of oxidation kinetic properties of coal on the spontaneous combustion in subsurface caved strata are studied. The process of oxygen chemisorptions on coal at different temperatures is measured by using the TG-DSC method and the properties for three coal samples are obtained. The coupled heat transfer model for porous media of coal and rock is established to simulate the thermal dynamical process in coal-bearing strata combined with the oxidation kinetic properties.

The evaluated heat evolution during oxygen chemisorptions and the kinetic parameters of three typical coals under different temperature levels illustrate that the coal oxidation is a multi-stage process separated by the critical temperature. The simulated results of the temperature distribution and the maximum combustion temperature due to the underground coal fire at different times show that the temperature of coal seam is nearly constant during a quiet long period after mining. This is caused by the slow oxidation process at low-temperature condition. Coal fire ignites and starts to penetrate to the up- and downward directions through the seam when the oxidation switches into the second stage after over the threshold temperature. The rank of coal has remarkable influence on the coal fire propagation.

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