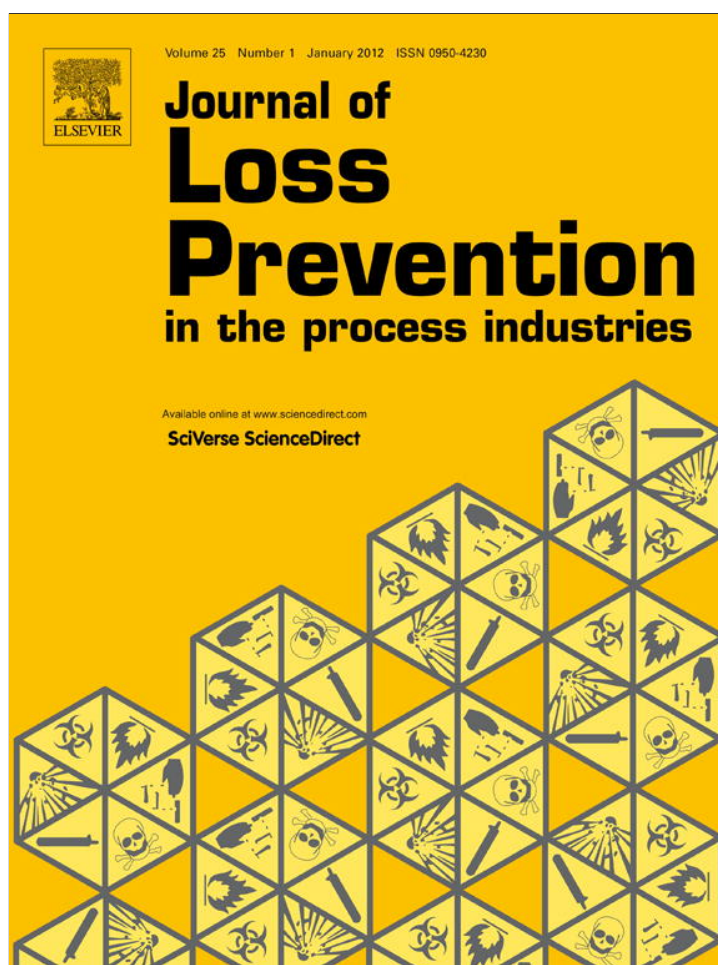




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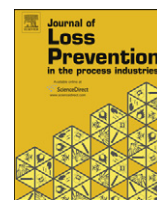
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The effect of ventilation on spontaneous heating of coal

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ABSTRACT

Ventilation plays an important role in the spontaneous heating of coal in an underground coal mine. If the ventilation rate is too high, heat is carried away by convection. If the ventilation rate is too low, the reaction rate becomes oxygen-limited. The effect of ventilation on the spontaneous heating of coal was investigated in an isothermal oven in this study. Experiments were conducted on three U.S. coal samples with ventilation rates ranging from 100 to 500 cm³/min. Experiments under ventilation were conducted to determine the critical ambient temperature, which is the minimum oven temperature required for a coal sample to achieve thermal runaway. Spontaneous heating tests were then conducted at various ventilation rates at the critical ambient temperature and the results were compared with spontaneous heating tests without ventilation. It was found that there is an optimum ventilation flow to produce the maximum rate of temperature rise at the critical ambient temperature. When the coal sample particle size was increased, a higher critical ambient temperature was required. The results in this study have application in the prevention of spontaneous combustion in underground coal mines.

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1. Introduction

Spontaneous heating has long been a problem in the mining, storage, and transport of coal. Spontaneous heating is a low temperature coal oxidation reaction which takes place when coal is exposed to air. If the heat produced by the coal oxidation is not adequately dissipated by conduction or convection, it results in a net temperature increase in the coal mass. Because coal oxidation is an irreversible exothermic reaction and its reaction rate increases with temperature, the increase in temperature leads to a higher oxidation rate. If not averted with appropriate action, this process results in thermal runaway and a fire ensues.

Much research has been done to understand the physical and chemical processes involved in spontaneous heating and the effects of different parameters on the spontaneous heating process (Akgun & Arisoy, 1994; Beamish & Arisoy, 2008; Carras & Young, 1994; Nugroho, McIntosh & Gibbs, 2000; Ren, Edwards & Clarke, 1999; Smith & Glasser, 2005; Smith, Miron & Lazzara, 1991; Wang, Dlugogorski & Kennedy, 2003). However, the chemical reaction between coal and oxygen at low temperatures is complex and still not well understood. Generally, three types of processes are believed to occur (Carras & Young, 1994): (i) physical adsorption of oxygen; (ii) chemical adsorption which leads to the formation

of coal–oxygen complexes and oxygenated carbon-species; and (iii) oxidation in which the coal and oxygen react with the release of gaseous products, typically carbon monoxide (CO), carbon dioxide (CO₂), and water vapor (H₂O).

In laboratory experiments, a number of methods has been used in attempting to predict the spontaneous combustion tendencies of coals, as reviewed by Wang, Dlugogorski and Kennedy (2006). In the U.S., the Bureau of Mines developed the minimum self-heating temperature (SHT) method using an adiabatic heating oven to evaluate the relative self-heating tendency to spontaneous combustion for U.S. coals (Smith & Lazzara, 1987). Although these evaluation methods can be useful for predicting the spontaneous heating potential of coal, there are difficulties in scaling the results to the large-scale conditions found in underground coal mines.

The spontaneous heating of coal in underground mines often occurs in mined-out areas (gobs) and is not easily detected. Computation fluid dynamics (CFD) modeling on spontaneous heating of coal in longwall gob areas has been conducted (Yuan & Smith, 2008). In mines that ventilate these mined-out areas, the amount of coal that accumulates in these areas and the degree of ventilation can combine to give optimum conditions for coal self-heating to occur. However, little research is available about the effect of ventilation on the spontaneous heating of coal. To better prevent the spontaneous combustion of coal in underground coal mines, it is important to understand the effect of ventilation on the reaction mechanisms of the low temperature oxidation of coal. In this paper, laboratory-scale experiments were conducted to investigate the effect of ventilation

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on spontaneous heating. The experimental results in this study provide insight into the interaction between ventilation and coal oxidation and improve the CFD modeling. The analysis of CO and CO₂ data will be presented in a separated paper.

2. Experimental procedure

Experiments were conducted in a programmable isothermal oven. The oven temperature can be held constant from 40 to 200 °C, within an accuracy of ± 0.5 °C. In each experiment, the oven is first turned on and preset at a certain temperature. After about 30 min, the isothermal condition is reached in the oven. Then, a stainless steel container with the coal sample is placed in the center of the oven. During testing, the coal sample is contained in a 6-cm diameter by 10-cm high brass wire mesh basket enclosed in a stainless steel container. Three 0.5-mm-diameter type K thermocouples are placed at the center of the coal sample, 3 cm above and below the coal center, respectively, to measure the coal temperatures. The ventilation gas—air or nitrogen—is introduced into the container through ports in the bottom and exits from the top, allowing the gas to flow through the coal sample. The gas is preheated to the oven temperature by being passed through heat exchange coils in the oven. Initially, the coal sample is heated to a selected temperature while exposed to a flow of dry nitrogen. When the coal sample reaches the oven temperature, the nitrogen flow is switched to the same flow rate of dry air. The exhaust gas exiting the container passes through a water trap before going through oxygen, carbon monoxide (CO), and carbon dioxide (CO₂) gas analyzers. The CO and CO₂ gas concentrations are measured continuously using an infrared gas analyzer, while the oxygen concentration is monitored continuously by a paramagnetic oxygen analyzer. Tests are terminated when the coal temperature begins to decrease or the temperature reaches 250 °C. A schematic of the experimental setup is shown in Fig. 1.

Coals from three U.S. mines were evaluated in these experiments. The proximate and ultimate analyses for the three coals are given in Table 1. The coal samples are freshly mined and shipped in airtight containers. To be sized for the experiment, the coal sample is first passed through a jaw crusher and then pulverized and sieved to the 100 $\times$ 200 mesh (75 $\times$ 150 μ m) size fraction. This coal is dried in the oven with a flow rate of 200 cm³/min nitrogen at 67 °C prior to the experiment. 150 g of coal sample is used in each experiment.

3. Experimental results and discussion

3.1. Determination of critical ambient temperature

Initial experiments were conducted to determine each coal's critical ambient temperature and activation energy, shown in Table 2.

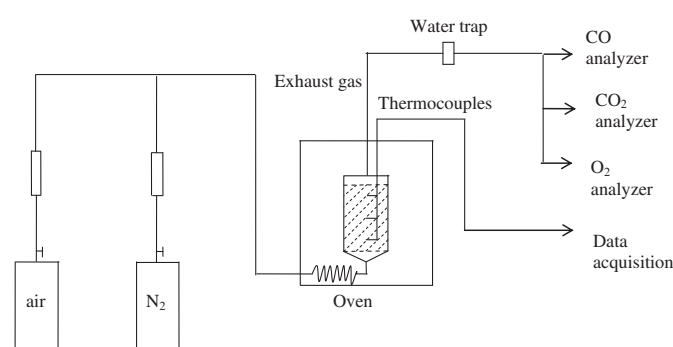


Fig. 1. Schematic of experimental setup and measurement.

Table 1

Proximate and ultimate analyses of the coal samples.^a

Coal sample	A	B	C
<i>Proximate analysis</i>			
Volatile matter (%)	36.88	38.40	44.03
Fixed carbon (%)	55.52	54.46	47.44
Ash (%)	7.60	7.14	8.53
<i>Ultimate analysis</i>			
Hydrogen (%)	5.52	5.83	5.47
Carbon (%)	81.88	82.72	77.79
Nitrogen (%)	1.49	1.57	1.48
Sulfur (%)	0.81	0.49	2.61
Oxygen (by difference)	10.30	9.39	12.65

^a Proximate analysis is on dry basis, and the ultimate analysis is on dry ash-free basis.

The critical ambient temperature is defined as the minimum oven temperature at which a thermal runaway is reached (Sujanti & Zhang, 1999). Here, thermal runaway is defined as a continuous temperature rise at the center of the coal sample. To determine the ambient temperature, experiments are conducted at an initial oven temperature with different airflow rates. If there is no thermal runaway, the oven temperature is increased by 5 degrees. Experiments are repeated until a thermal runaway is reached. This minimum oven temperature at which the thermal runaway occurs is the critical ambient temperature for the coal sample at this flow rate. Experiments were conducted on coal sample A at an initial oven temperature of 95 °C with airflow rates ranging from 100 to 500 cm³/min. Eventually, a thermal runaway was reached for the sample A with the oven temperature at 110 °C at an airflow rate of 300 cm³/min. With this oven temperature at the airflow rates of 100 and 200 cm³/min, no thermal runaway occurred. With this oven temperature at the airflow rate of 500 cm³/min, the thermal runaway also occurred. Similar tests were conducted for coal samples B and C. For coal sample B, thermal runaway occurred at an oven temperature of 115 °C and an airflow rate of 200 cm³/min. This critical ambient temperature is slightly higher than the critical ambient temperature for sample A. Tests on coal sample C resulted in a critical ambient temperature of 100 °C with the airflow rate at 200 cm³/min or greater.

The measurement of the coal's activation energy is based on the heat release method (Jones, Chiz, Koh & Matthew, 1996) using the same isothermal oven. In this method, coal samples were placed in a 10-cm high, open-topped cubical brass wire mesh basket. This was placed in the oven without any airflow. Two thermocouples were placed at the center and at half the distance between the center and the surface of the sample. When the temperatures recorded at two thermocouples are equal, the thermal conduction within the sample becomes zero. Therefore, the one-dimensional energy balance for the self-heating for the sample becomes

$$\left(\frac{\partial T}{\partial t}\right)_{T_0} = \frac{kQ}{C_p} \exp\left(-\frac{E}{RT_0}\right)$$

where T is the sample temperature, t is the time, k is the pre-exponential factor, Q is the heat of reaction, C_p is the specific heat

Table 2

Activation energy and SHT of coal samples.

Coal sample	Critical ambient temperature, °C	Measured activation energy (kcal/mol)	Calculated SHT, °C
A	110	22.3	72
B	115	20.2	78
C	100	9.5	57

capacity, E is the activation energy, R is the gas constant and T_0 is the temperature at the point when the temperatures from two thermocouples cross. This equation enables the estimation of the kinetics of the oxidation reactions. Plotting the logarithm of the transient heating $\ln(\partial T/\partial t)_{T_0}$ at the temperature T_0 versus the reciprocal temperature $1/T_0$, the activation energy can be determined from the slope of the resulting straight line. This method has been demonstrated to produce comparable results as other methods (Malow & Krause, 2004).

Also shown in Table 2 is each coal's minimum self-heating temperature (SHT). The SHT is a measure of a coal's reactivity, and is calculated based on the method established by Smith and Lazzara (1987) using coal analysis data. The differences in the apparent relative reactivity between the three coal samples were also reflected in their different SHTs. Based on calculated SHT data, sample C is the most reactive coal, while sample B is the least reactive coal.

3.2. Effect of ventilation rate

Fig. 2 shows the temperature-time and exit oxygen concentration histories for the test of coal sample A at its critical ambient temperature of 110 °C at different locations under the airflow rate of 300 cm³/min. The bottom temperature exhibited a clear three-stage increase. The first stage is a slow increase, the second one is a fast increase, and the third one is a slow increase again. The center temperature decreased slightly but increased slowly again in the third stage. The top temperature was much lower than the center and bottom temperatures. The center temperature increased slightly faster than the bottom in the first stage, while the bottom temperature increased faster than the center in the second stages, probably because not enough oxygen was available at the center.

Fig. 3 shows the center coal temperatures of coal sample A under different ventilation rates at the critical ambient temperature of 110 °C. With ventilation rates of 100 and 200 cm³/min, the center temperatures first increased then decreased. Under these conditions, no thermal runaway was reached. Although a thermal runaway was reached with the flow rates of 300 and 500 cm³/min, the center coal temperature decreased a little, then increased very slowly after reaching over 200 °C. To better understand the effect of ventilation on spontaneous heating, an experiment was then conducted for coal sample A without any ventilation, designated as “in open”. During the test, a coal sample was first placed in the same brass wire mesh basket. The basket was then put into the oven without being enclosed in the stainless steel container. The oven

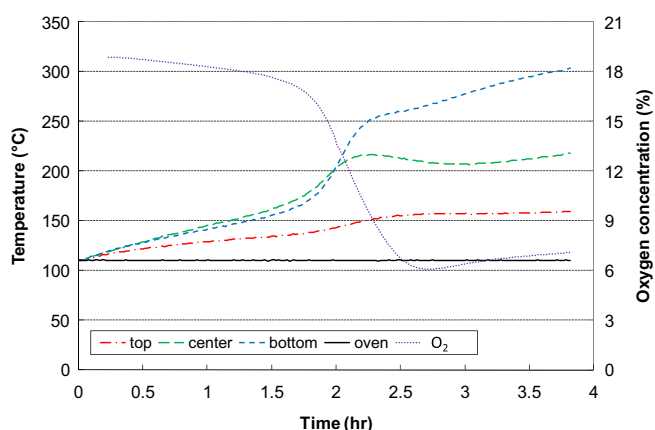


Fig. 2. Coal temperatures at different locations and oxygen concentration under the airflow rate of 300 cm³/min for sample A.

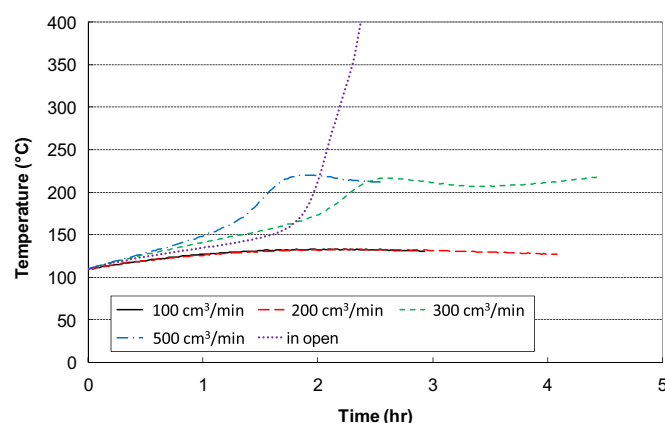


Fig. 3. Center coal temperatures under different ventilation rates with the oven temperature at 110 °C for sample A.

temperature was first set at 90 °C and the center coal temperature was monitored continuously. If there was no thermal runaway at a given oven temperature, the oven temperature was increased by 5 °C. Tests were repeated until a thermal runaway was reached. When the oven temperature was below 120 °C, coal temperatures always increased first, then decreased slowly. When the oven temperature was at 120 °C, a thermal runaway was reached. This temperature was defined as the self-ignition temperature (SIT) for combustible materials by Krause and Schmidt (2001). This SIT was higher than the critical ambient temperature, 110 °C for coal sample A, at which the thermal runaway was reached with the flow rates of 300 and 500 cm³/min, indicating that with the forced ventilation, the coal sample could reach a thermal runaway at a lower temperature than its self-ignition temperature.

The center coal temperature without ventilation was also compared with the center coal temperatures with ventilation in Fig. 3. It is interesting to note that without ventilation, the center coal temperature exhibited a two-stage increase. The first stage was a slow increase, followed by a fast increase. Compared with center coal temperatures with ventilation, the significant difference is that the center coal temperature without ventilation increased over 400 °C very quickly in the second stage. However, the center coal temperature with ventilation increased faster in the first stage than that without ventilation, indicating that ventilation might have enhanced the spontaneous heating at this stage. The temperature increase was then much slower than that without ventilation. In the third stage, the temperature increase became nearly flat. Table 3 lists the rates of temperature rise for coal sample A under different ventilation rates. It can be seen that the rate of temperature increase in the second stage “in open” was 3–4 times faster than the temperature increase with ventilation. This is likely due to the fact that under ventilation conditions the amount of oxygen in the airflow was not enough to sustain the fast coal oxidation reaction in the late stage, and the temperature rise became ventilation controlled; while without ventilation, the coal sample was exposed to the ambient air, and enough oxygen was available for the coal oxidation reaction.

Table 3

Rate of temperature increase for coal sample A (°C/h) at 110 °C ambient temperature.

Ventilation rate	1st stage	2nd stage	3rd stage
300 cm ³ /min	28	109	16.7
500 cm ³ /min	40	140	—
“In open”	21.7	466.7	—

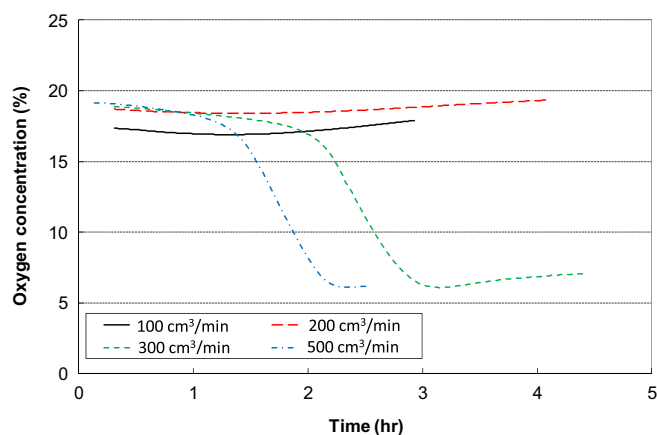


Fig. 4. Oxygen concentration at the exit of the oven under different ventilation rates for sample A.

Fig. 4 shows the oxygen concentration at the exit of the oven under different ventilation rates for sample A. With the airflow rate of 300 and 500 cm³/min, the oxygen concentration also exhibited three stages. In the first stage, the oxygen consumption rate was small, while it became significantly larger in the second stage. However, in the third stage, the oxygen concentration actually increased slowly. With the airflow rate below 300 cm³/min, the oxygen concentration only decreased slowly at the beginning of the first stage, but then increased without experiencing a fast decrease stage.

Fig. 5 shows the center temperatures for coal sample B at its critical ambient temperature of 115 °C under different ventilation rates. As the airflow rate increased, the rate of temperature rise in the first stage also increased. The oxygen concentrations at exit under different ventilation rates for this test are shown in Fig. 6. Compared to Fig. 4, sample B had a higher oxygen consumption rate (reaction rate) in the first stage than sample A at the same airflow rate, probably because of its lower activation energy.

Fig. 7 shows center coal temperatures for sample C under different ventilation rates at its critical ambient temperature of 100 °C. At this oven temperature, thermal runaway was reached with the airflow rate of 200 cm³/min or greater. Fig. 8 shows the oxygen concentrations at the exit for coal sample C under the different ventilation rates. It is clear that at the flow rate of 100 cm³/min, thermal runaway did not occur, even though the

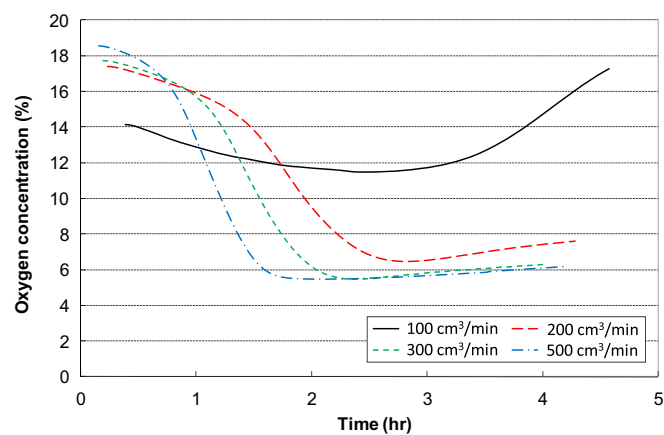


Fig. 6. Oxygen concentration at the exit of the oven under different ventilation rates for sample B.

lowest oxygen concentration was below 6%. It should be pointed out that the oxygen concentration for sample C was lower at 200 cm³/min than at 300 cm³/min, which is different from samples A and B. This is probably because sample C already adsorbed much more oxygen initially (oxygen concentration at 13.2%) at 200 cm³/min than sample A and B (oxygen concentrations: 18.7% and 17.4%, respectively). Therefore, at the higher flow rate of 300 cm³/min, the oxygen adsorbed by samples A and B increased significantly, while it did not change significantly for sample C. This led to a higher oxygen concentration for sample C at 300 cm³/min. At 500 cm³/min, the oxygen adsorbed by sample C increased significantly as it did with samples A and B.

3.3. Effect of coal properties

Fig. 9 compares the center coal temperature rise for three samples under the airflow rate of 300 cm³/min. The oxygen concentrations for the three samples are compared in Fig. 10. For the center coal temperature rise, sample A clearly exhibited a three-stage temperature increase with the slowest increase rate in the first stage. Sample C exhibited a two-stage temperature increase with the fastest increase rate in the first stage. The oxygen concentrations in Fig. 10 show that the lowest oxygen concentration was reached for coal sample C, implying that for the more reactive coal, the smaller lowest oxygen concentration is required

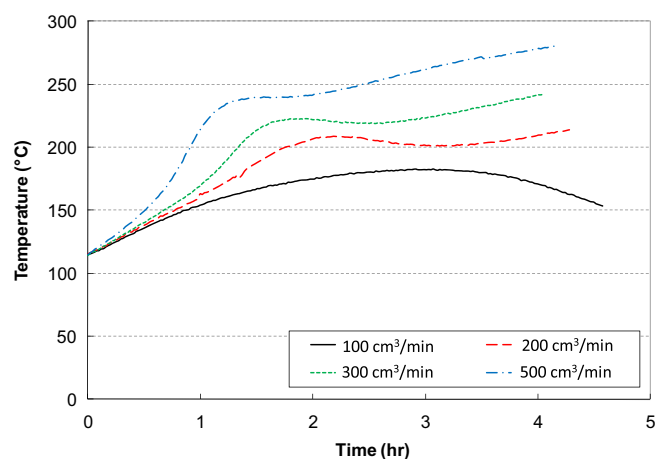


Fig. 5. Center coal temperatures under different ventilation rates with the oven temperature at 115 °C for sample B.

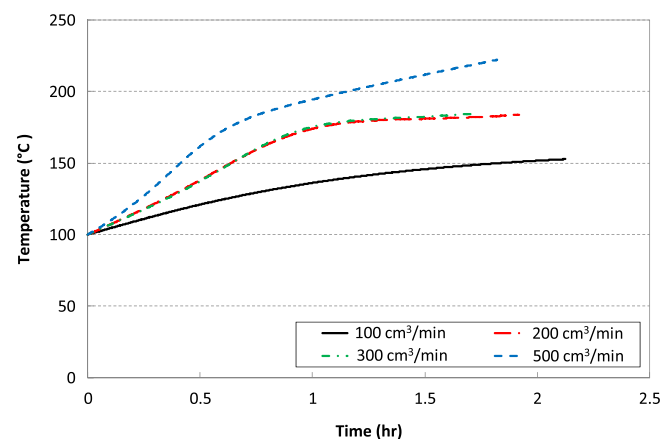


Fig. 7. Center coal temperatures under different ventilation rates with the oven temperature at 100 °C for sample C.

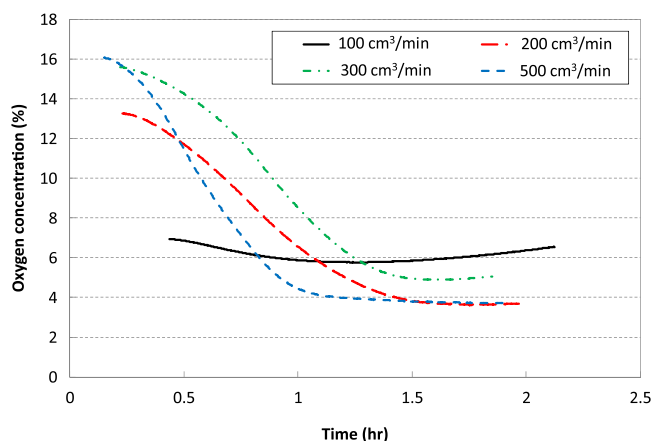


Fig. 8. Oxygen concentration at the exit of the oven under different ventilation rates for sample C.

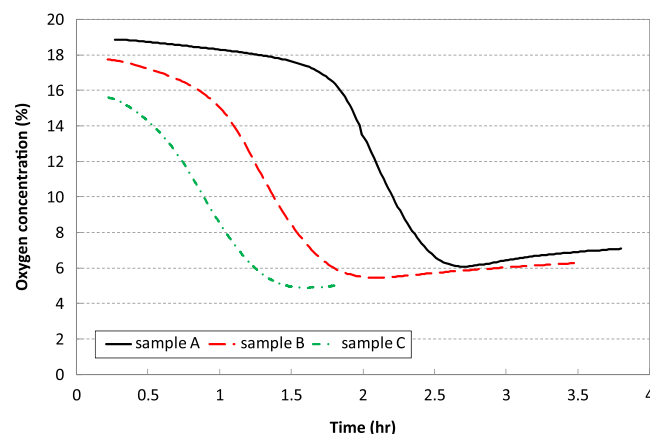


Fig. 10. Oxygen concentrations for three samples under the airflow rate of 300 cm³/min.

to sustain the coal oxidation. Compared to the activation energy and SHT data in Table 2, the experimental results for the three coal samples also indicate that the critical ambient temperature is related to the SHT, while the oxidation rate, especially in the first stage, is related to the activation energy of the coal. Therefore, for a certain type of coal to reach a thermal runaway under ventilation, the reaction rate in the first stage must be higher than that “in open”.

It is interesting to note that no thermal runaway was reached for all three coal samples with the airflow rate of 100 cm³/min, indicating that there exists, for ambient temperatures lower than the SIT, a minimum ventilation rate below which a thermal runaway can be prevented. Although no thermal runaway was achieved at this airflow rate, the oxygen concentrations were still reduced at exit because of physical and chemical adsorption. The exit oxygen concentrations at 0.5 h for coal samples A, B, and C were 17.2%, 14.0% and 6.9%, respectively. These concentrations imply that a more reactive coal adsorbs more oxygen and that the amount of oxygen adsorbed initially determines whether a thermal runaway can be reached. If enough oxygen is adsorbed—as in the cases for sample B and C with the airflow rate of 200 cm³/min or above, and in the case for sample A with the airflow rate of 300 cm³/min or above—then thermal runaway could be reached. Thus, the amount of oxygen adsorbed by a certain coal mass will depend not only the physical and chemical properties of the coal, but also the oven temperature (ambient temperature).

3.4. Effect of ventilation with initial coal temperature below the critical ambient temperature

It is important to detect spontaneous combustion in its early stages, so that appropriate measures can be taken to prevent the spontaneous heating from developing into a fire. Therefore, experiments were conducted to investigate the effect of ventilation on the spontaneous heating with the oven temperature below the critical ambient temperature of the coal sample. Coal sample C was used in the experiments, and the oven temperature was set at 90 °C. At this temperature, a thermal runaway could not be reached. The center coal temperatures and oxygen concentrations at the exit at this oven temperature under different ventilation flow rates are shown in Figs. 11 and 12. With the oven temperature at 90 °C, the maximum temperature rise, about 48 °C, was reached at the flow rate of 200 cm³/min, as shown in Fig. 11. The maximum temperature at the flow rates of 300 and 500 cm³/min are nearly the same, but lower than that at the rate of 100 cm³/min. This is significantly different from the case with the oven temperature at 100 °C, as shown in Fig. 7, in which the coal temperature increased with the airflow rate. Oxygen concentrations shown in Fig. 12 clearly show that no significant chemical reaction took place. At this oven temperature, no thermal runaway occurred at any airflow rate. Therefore, oxygen was likely consumed by chemical adsorption and low-rate oxidation reaction. Although not much oxygen was

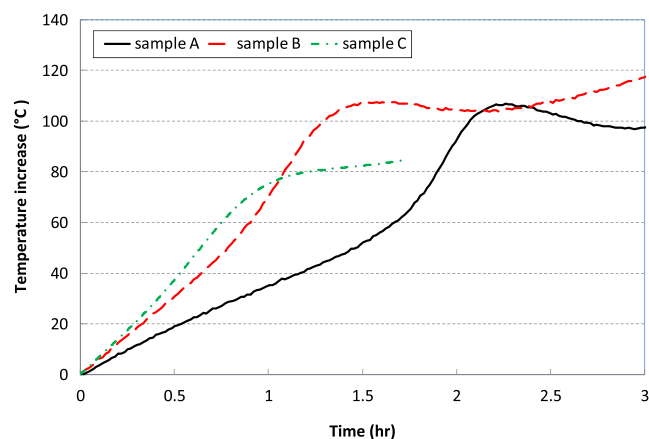


Fig. 9. Center coal temperature rises for three samples under the airflow rate of 300 cm³/min.

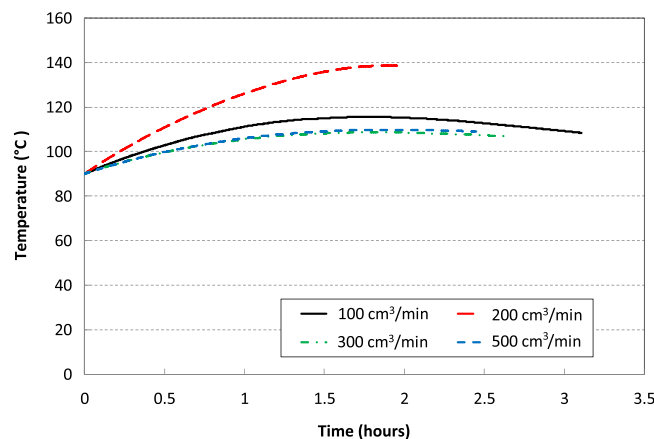


Fig. 11. Center coal temperatures under different ventilation rates with the oven temperature at 90 °C for sample C.

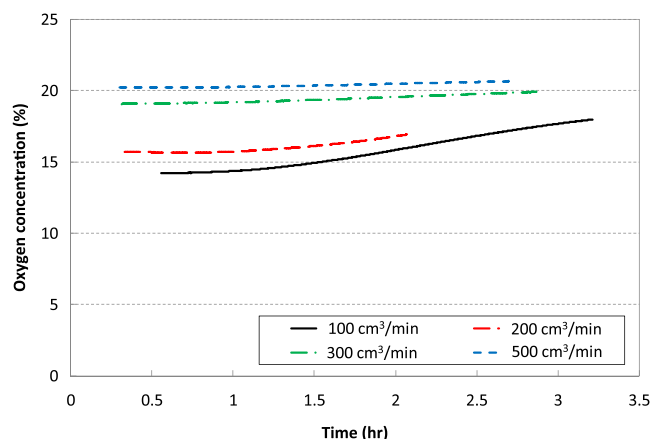


Fig. 12. Oxygen concentration at the exit of the oven under different ventilation rates for sample C.

consumed, the oxygen concentration at the exit was still reduced to about 14% at the airflow rate of 100 cm³/min because the amount of oxygen at this flow rate is limited. However, at the airflow rate of 500 cm³/min, the oxygen concentration at the exit was still above 20% because five times the amount of oxygen was available at this flow rate. These results indicate that with a low ventilation rate, 100 cm³/min, less oxygen was available, leading to less heat generated. With a higher ventilation rate, 200 cm³/min, more heat was generated, which led to a higher coal temperature. However, as the ventilation was further increased to 300 and 500 cm³/min, the larger air velocity took away more heat, leading to the lower coal temperature. Therefore, the flow rate of 200 cm³/min generated the highest coal temperature. These results indicate that ventilation plays a different role in relation to spontaneous combustion when the initial coal temperature is below the critical ambient temperature.

3.5. Effect of coal particle size

Tests were conducted with a larger size coal sample A, 150 by 300 μm, to examine the effect of coal particle size on spontaneous heating under different ventilation rates. The initial oven temperature was set at 110 °C, the critical ambient temperature found for this coal using 75 by 150 μm particles. When there was no thermal runaway, the test was repeated with the oven temperature

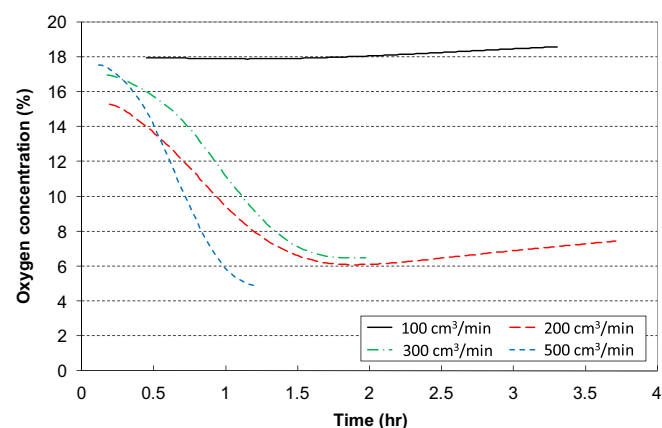


Fig. 14. Oxygen concentration under different ventilation rates for larger size sample A.

increased in 5-degree increments. The thermal runaway was reached when the oven temperature reached 130 °C, indicating that a higher critical ambient temperature is required for a larger size particle.

Fig. 13 shows the center coal temperatures under different ventilation rates. It is interesting to note that the temperature rise exhibited a two-stage increase, compared to the three-stage increase shown in Fig. 3. With the increased coal particle size, the available coal surface area was reduced. Therefore, less oxygen could be adsorbed by the coal surface. In order to adsorb enough oxygen to increase the temperature high enough to support a fast oxidation rate for the thermal runaway, the higher initial temperature is required. However, at the higher temperature, the effect of kinetics becomes more significant. Even with the ventilation rate of 200 cm³/min, thermal runaway was reached.

Fig. 14 shows the oxygen concentration at the exit of the oven under different ventilation rates. The oxygen concentration was also lower at 200 cm³/min than that at 300 cm³/min. As discussed above for Fig. 8, this is because enough oxygen was already adsorbed at 200 cm³/min, probably due to the higher ambient temperature. The oxygen concentration did not increase significantly at 300 cm³/min.

4. Conclusions

Experiments were conducted in an isothermal oven to investigate the effect of ventilation on spontaneous heating of coal. Experimental results demonstrate that a thermal runaway could be reached at a lower temperature with enough ventilation airflow than “in open” (without ventilation). With enough ventilation, the rate of coal temperature rise in the first stage of spontaneous heating was higher; however, the rate was slower in the second stage compared to that without ventilation. In the third stage, the temperature and oxygen concentration both increased slowly, indicating that it may take a much longer time to develop a fire compared to the unventilated state. As the airflow rate increased, the rate of temperature rise in the first stage also increased.

Experimental results also indicate that the critical ambient temperature at which a thermal runaway can be reached with enough ventilation is related to the SHT of coal, while the oxidation rate, especially in the first stage, is related to the activation energy of the coal. For a certain type of coal, whether a thermal runaway could be reached depends on the reaction rate in the first stage. For the more reactive coal, the minimum oxygen concentration required to sustain the coal oxidation was lower.

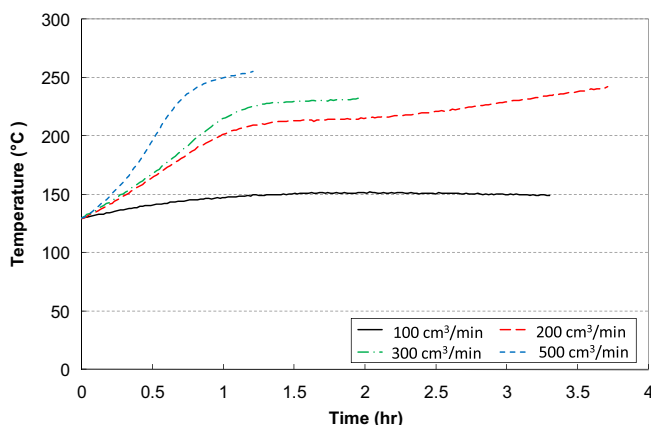


Fig. 13. Center temperatures under different ventilation rates for larger size sample A with oven temperature at 130 °C.

For this experiment, when the initial coal temperature was below the critical ambient temperature, the highest temperature was reached with a moderate ventilation rate. The lower ventilation rate would not supply enough oxygen, while the higher ventilation rate carried away more heat, both leading to a lower temperature. For the larger-sized coal sample A, 150 by 300 μm , a higher critical ambient temperature was required to reach thermal runaway.

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