A Technique for Decreasing Reactivity of Coal Material to Suppress the Oxygen Absorption Process

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Abstract. The paper describes the mechanisms of self-ignition formation in coal liable to spontaneous combustion, on the basis of experimental works performed to analyze heat and mass transfer in the coal-air system. A new approach was developed to the coal self-heating suppression and thermodynamic control of the oxidation process. The influence of coal moisture content and thermal behaviour of air in the cooling process was studied during moisture evaporation.

1. Introduction

For establishing the determining factors that favour oxidation and self-heating of coal, extensive studies were conducted to analyze the influence of combined parameters (rate of flow, temperature, humidity, oxygen concentration) of air on reactivity of coal material, which has various natural moisture contents, in a laboratory environment and underground [1-5].

It has been experimentally proved that self-heating of coal (up to 80 °C) occurs at a relatively low rate of air filtration \((3-7) \times 10^{-3} \text{ m/s}\), and the determining factor is its moisture exchange in the heterogeneous coal-air system, which is dependent on aerodynamic and thermo physical properties of air, and moisture content of coal. At relative humidity being constant, the oxidative process and self-heating of coal rapidly slow down, accompanied with a simultaneous decrease in rate, temperature and concentration of oxygen in the intake ventilation air flow. However, these parameters, used to quantify any ventilation loss underground in order to prevent spontaneous combustion of coal in a goaf at working areas, are difficult to control. Therefore, decelerating the oxidation process by increasing a relative humidity up to 100 % at the constant rate and oxygen concentration has a practical importance. The physical nature of the effect that results in inactivating coal compaction is associated with changing the direction of moister exchange in the coal-air system, thereby reducing the rate of drying and chemical activity of coal.

According to the studies conducted for this purpose, an increase in coal moisture decreases its ability to absorb oxygen, with the slow down effect on the coal oxidation taking place for a long time, what is important for underground operations. Earlier researches established that, at the initial phase of
oxidation, the moist coal absorbs less oxygen than the dry coal (Figure 1). However, the amount of oxygen absorbed by the moist and dry coals becomes equal after 144 hours.

![Graph showing the time dependence of the amount of oxygen absorbed by coal](image)

The previously watered and then dried coal becomes more active to oxygen. The activating effect of water that is removed from coal by drying can be explained by the increased availability for coal-derived substances to reach both methane and oxygen. It is known that, in all cases, the residual content of methane in the preliminarily watered coal is 30-50 % higher than that of the un-watered. The absorbable by coal methane behaves as if it screens a highly developed surface, prevents oxygen molecules to connect with coal particles. Besides, the higher moisture of coal results in increasing its thermal parameters that accelerates a dissipation of heat released due to oxidation and oxygen adsorption, and inhibits the development of an autocatalytic process of oxidation and self-heating of coal.

2. Results and Discussion

On the basis of theoretical and large-scale laboratory studies relating to heat and mass transfer in the coal-air system [6], it has been established that the mechanisms of self-ignition formed in the coal susceptible to spontaneous combustion, allowing for real-time factors, are as follows:

- Density of the resulting moisture flow (from coal to air) and heat released during coal oxidation have an identical progressive pattern over time in reference to the rates of air filtration through the compact coal in the range of \((2.5-4.6)\times10^{-3}\), with corresponding to both laminar and transitional modes;

- At a coal temperature of 32-45 °C and a rate of air filtration \(4.7-6.0)\times10^{-3}\) m/s, the decrease in moisture exchange takes place that results in initiating thermodynamic stabilization of the heterogeneous oxidation reaction in the coal compaction due to the deepening of evaporation area inside lumps of coal and the increasing of a moisture diffusion pathway;
- Lowering a temperature of the intake air by 3-5 °C and its relative humidity down to 70 % at the second phase of intensive moisture evaporation from coal leads to an extremum in the coal-air system shifting to the zone with a higher risk of occurring spontaneous coal combustion processes;

- The self-heating process in the coal compaction changes into the self-ignition process when the coal moisture content is decreased by on average 60%, if compared to the natural bed moisture, under the turbulent air flow conditions.

The mechanisms established in forming places of spontaneous coal combustion allowed the investigators to come to a scientifically based conclusion that, during the self-heating phase, the moisture of coal is the most important parameter in determining this phase duration. In this context, a new approach was developed to the spontaneous combustion inhibition to improve thermodynamic stability of the oxidation process by controlling this parameter.

This approach relates to maintaining a thermodynamic state of the heterogeneous coal-air system within a rigidly fixed range, and shifting its extremum to the area with a low propensity for spontaneous coal combustion by means of changing the orientation of moisture exchange processes between the phases in this system with a purpose of regulating thermal parameters of air by using artificial or natural aerosols.

It is known that any rank of coal has the so-called critical moisture content of coal, and when this content is reached, the rate of coal oxidation sharply increases by 1.5-2 times. It is established for coals to have a critical moisture content varying from 2 or 3 % to 17-18 % for brown coals. Thus, the results of theoretical and laboratory studies, aimed at identifying a relationship between critical moisture of coal and distribution of moisture in the micro- and super microporous structure of coal, can be used to describe mechanisms of oxidation reactions in coal as well as calculate temperatures of coal self-heating.

To date, there are a considerable number of mathematical models and software developed and employed to simulate coal self-heating processes [7]. Some of them allow for either the time dependence of absorption rate or an increase in the rate of oxygen absorption by coal at critical moisture contents. At the same time, experimental studies prove the combined influence of these factors on the process of coal self-heating. In this regard, it is relevant to design a procedure for the calculation of temperatures in coal compactions to improve the existing models and take into account both the processes of deactivating coal and impacts associated with the critical moisture content of coal.

A number of methods and techniques are known that used for quantification of thermal behaviour of coal while coal self-heating and various natural and man-induced factors, having an influence on this behaviour. Those methods served as the basis for the development of a technique to be used for preventing spontaneous coal combustion.

However, there is currently no procedure that used to analyze the combined impact of coal deactivation depending on time and coal activation at critical moisture content on the thermal behaviour of the self-heating coal. The work in [8] describes the procedure for quantifying the thermal behaviour of the self-heating coal, allowing for the coal deactivation over time and higher rates of coal oxygen absorption, with a moisture loss through super micropores.

According to the research of V.M. Maevskaya, there are three phases of the spontaneous coal combustion process: heating, moisture evaporation, active oxidation. In general, the thermal behaviour modeling verifies the characteristics peculiar to each phase of spontaneous combustion. However, it should be noted that during the moisture evaporation phase the temperature decreases due to coal deactivation over time, not intensive moisture evaporation.

The phase of active oxidation starts with the moisture loss through super micropores.

The tendency towards a thermodynamic equilibrium at constant environmental conditions is characteristic of thermodynamic systems, including the heterogeneous coal-air system. With regard to the issues related to the spontaneous combustion prevention in coal, the problem of regulating these processes reduces to the explanation of the mechanism and establishment of parameters ensuring the equilibrium state with decreasing a temperature in self-ignition sites.
Coal Moisture is the first and most important parameter that influence the duration of coal self-heating to a stabilized temperature in the coal compaction. Therefore, to improve thermal stability in the places propitious for coal self-heating and self-ignition, it is recommended to influence the concentration of oxygen and the rate of air filtration in mined-out areas [9].

The significant slowing down of self-ignition in the oxidizing material can be also achieved by reducing the initial temperature of interacting components [10]. In this case, the duration of low-temperature oxidation increases. Simultaneously, according to the Arrhenius equation, a decrease in temperature causes the reduction of reaction rate in oxidation. Therefore, coal cooling also reduces the amount of heat released that will raise potential to prevent self-ignition from occurring and spontaneous coal fires.

Coal temperature in the worked-out area can be lowered by supplying low-temperature medium such as cooling gas. However, gas compositions have a low density and specific heat content, so large volumes of gas are required for cooling the coal. Cooling medium consumption can be significantly reduced by adding particles of a frozen fluid to the gas [11,12]. The intensity of heat removal from coal and rocks increases dramatically due to the absorption of heat during phase transition, as well as higher density and specific heat of fluid particles. It is recommended to use nitrogen as cooling gas, as it does not interact with carbon.

For an effective formulation designed to suppress spontaneous coal combustion. It is important to investigate how the coal moisture and air thermal parameters influence the cooling process and its intensity, when moisture evaporates from the surface, and determine how wetting fluids penetrate the coal mass. The following experiment was conducted for the purpose.

The crushed coal in an amount of 1 kg was poured into a container in the form of a funnel (Figure 2), the air that has various intensity rates and humidity was supplied from the bottom. The outlet air temperature, flow rate and humidity were then measured. Samples of coal with natural bed moisture and samples with a 10 % and 20 % increase in moisture content were used in the course of experiment.

For the first series of experiments, the samples of coal with natural bed moisture were used under the following conditions: the rate of air flow was 65 l/min; the temperature of supply air was 25 °C; the air humidity was 20 %. The temperature was measured every 5 minutes.

According to the results obtained in the course of experiments, the temperature of the coal watered by 10 % decreased by 3 °C during the period of 15 minutes, and watered by 20 % decreased by 4 °C (Figure 3). Consequently, the mechanism can be assumed to be evident: when the coal compacted around worked-out areas is watered to have a higher content of moisture, then blown with dry air, the decrease of coal temperature can be achieved down to a safe value. This can be implemented by periodic water spraying into the air flow at the worked-out area. As a result, coal and rocks become wet. After injecting the fluid into the mined-out area, it is necessary to supply a dry gas, such as nitrogen.

Figure 2 – An installation used for determining the rate of evaporation in the coal compaction: 1 – Compacted and crushed coal; 2 – Air supplied for blowing (of various humidity); 3 – A temperature and humidity meter; 4 – An air flow meter
Thus, it can be concluded that the deactivation of coal over time is an important parameter in modeling self-heating of coal which makes the simulation process more accurate. It is established in a laboratory environment, when the coal compaction is blown with absolutely dry gas and the humidity of air is high, a decrease in coal temperature is not observed for both cases with using dry and wetted air.

3. Conclusions

The blown dry gas makes particles of previously injected fluid evaporate more actively that result in a decrease in the coal temperature.

The preliminarily watered and then dried coal becomes more active to oxygen. A higher moisture content of coal promotes the increase of its thermal parameters that contribute to more rapid dissipation of heat released in the oxidation process due to oxygen absorption, and suppress the progress of autocatalytic oxidation process and coal self-heating.

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