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Kinetics characteristics of coal low-temperature oxidation in oxygen-depleted air

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ABSTRACT

The longwall gob (mined-out) area is one of the main places that are prone to coal spontaneous combustion and most of the residual coal in it is in oxygen-depleted air as it is a semi-enclosed space. A DSC (Differential scanning calorimetry) test system was designed to accurately test the heat generation of coal oxidation in different oxygen concentration atmosphere, based on which the kinetics parameters (activation energy and pre-exponential factor) of coal low-temperature oxidation in oxygen-depleted air were solved out. The results show that the kinetics parameters present obvious stage features and the lower the oxygen concentration is, the smaller is the difference of the kinetics parameters that in different oxidation stages. When the oxygen concentration is lower than 5% and 3% for jet coal and meagre coal respectively, the kinetics parameters of slow oxidation start to be greater than that of rapid oxidation. Both in the slow oxidation and rapid oxidation stage, with the decrease of oxygen concentration, kinetics parameters present significant decline on the whole while in different oxygen concentration range, the decline rate is different. It's concluded that when assessing the residual coal's self-heating risk, we need to use the corresponding kinetics parameters of coal oxidation in the oxygen concentration of the location where the residual coal is and the safety factor will be greater to only use the kinetics parameters of coal oxidation in slow oxidation stage. This study is of great significance for the assessment and control of the self-heating risk of coal that in different oxygen concentration atmospheres of the longwall gob areas.

1. Introduction

The majority of fires in coal mines are caused by spontaneous combustion of coal, and the longwall gob area is one of the main places that are prone to coal spontaneous combustion (Li, 1998; Taraba and Michalec, 2011). Fires occurred in the longwall gob areas will not only produce large amounts of toxic and harmful gases, a serious threat to the life of staff, and may also lead to other major accidents like gas explosion. Therefore, the prevention of spontaneous combustion of residual coal in longwall gob areas has always been an important task during mining.

The longwall gob area is a semi-enclosed space (Shao et al., 2015), and due to the oxygen consumption of residual coal oxidation, inadequate supply of fresh air and the dilution effect of gas released by residual coal, the oxygen concentration decreases in longwall gob areas with the increase of distance from the working face. Meanwhile, in the mining process, to prevent the low-temperature oxidation of residual coal, many mines continue to inject nitrogen to longwall gob areas for the further reduction of the oxygen concentration (Mohalik et al., 2005; Ray and Singh, 2007). So, in the longwall gob areas, most of the residual coal is in oxygen-depleted air, where the oxygen concentration is less than 20.96%. But, so far, because of the unclear understanding of the self-heating risk of coal in different oxygen concentration atmospheres, it is difficult to determine the effective area of the injected nitrogen where it plays a role and identify the hazardous areas where residual coal is prone to spontaneous combustion.

Many scholars have carried in-depth researches on how to assess the self-heating risk of coal. A series of studies focused on

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ranking the susceptibility of coal to spontaneous combustion by using different laboratory methods, such as adiabatic oxidation method (Vance et al., 1996; Ren et al., 1999; Beamish et al., 2000, 2001), activation energy method (Liu et al., 1999; Semsogut and Cinar, 2000), heat release method (Jones et al., 1996, 1998, 1999), ignition temperatures (Herbig and Jess, 2002), crossing point temperature (CPT) method (Bagchi, 1965; Nandy et al., 1972; Sujanti et al., 1999; Mahidin et al., 2002) etc.; Large scale spontaneous combustion experimental apparatus have also been developed by many researchers to obtain the low temperature oxidation law of coal for the prediction of coal spontaneous combustion (Smith, 1991; Chen and Stott, 1997; David Cliff et al., 1998; Deng et al., 1999). Meanwhile, a lot of papers have been published on the topic of the assessment of the self-heating risk of coal by simulation coal spontaneous combustion process through developing one-, two- or three-dimensional mathematical model of coal self-heating. (Srinivasan Krishnaswamy et al., 1996; Akgun and Essenhigh, 2001; Fierro et al., 2001; Yuan Liming and Smith, 2008; Taraba and Michalec, 2011; Zhu et al., 2013). Many other methods for the classification of coal with respect to spontaneous combustion susceptibility, such as neural network approach (Panigrahi and Sahu, 2004), kinetics based simulation method (Qi et al., 2014), Fuzzy c-means clustering approach (Sahu et al., 2012) have also been discussed. However, these studies mainly regard the coal that is in air atmosphere as the research object. Currently, few studies can be found that focused on the self-heating risk of coal under oxygen-depleted air.

For assessing the self-heating risk of coal, the accurate solution of the kinetics parameters (activation energy and pre-exponential factor) of coal oxidation is needed. Activation energy and pre-exponential factor, main characterization parameters for thermal hazard of substances, are the main factors in determining the reaction rate (Li et al., 2014). The application of Semenov model, Frank-Kamenetskii model and Thomas model, three existing mature theoretical model for the assessment of thermal hazard of substances, and the numerically simulation of low-temperature oxidation of coal by developing mathematical model, are both based on the solution of kinetics parameters. Many scholars (Kok and Okandan, 1994, 1996; Jones et al., 1998; Li et al., 2014) have conducted kinetics analysis of coal oxidation, however, most of them regarded the entire oxidation process of coal from ambient temperature to about 600 °C as the research object and focused on the high-temperature phase. The kinetics parameters obtained in these studies mainly reflect the kinetics characteristics of coal at combustion phase rather than at low-temperature oxidation phase. At the same time, very few papers have been published about kinetic analysis of coal under different oxygen concentration atmospheres. As the prevention of coal spontaneous combustion is actually the inhibition of coal's low-temperature oxidation phase, it is important to perform further studies to well understand the kinetics characteristics of coal during low-temperature oxidation in oxygen-depleted air.

The purpose of this investigation is to determine the kinetics characteristics of coal low-temperature oxidation in oxygen-depleted air. A DSC experimental system was designed for simulating coal oxidation under different oxygen concentration atmospheres and testing its heat generation. The variation regularity of kinetics characteristics of coal oxidation in oxygen-depleted air was obtained based on the thermal characteristics as exothermic oxidation is the root cause of coal spontaneous combustion. The outline of the paper is as follows: Section 2 shows the basic information of the coal sample and describes the test system and experimental protocols; Section 3 includes the heat release behavior, the method for preliminary data treatment, as well as the kinetics characteristics of coal low-temperature oxidation and the discussion about its application; Section 4 is the conclusions of this study.

2. Experimental section

2.1. Coal samples

Two experimental samples of different rank (one jet coal and one meagre coal) were collected in this study. The fresh coal samples were tightly wrapped using plastic wrap in sampling sites and were transported to the laboratory as soon as possible. Then, after removing the surface coal, the coal samples were ground in a glovebox under inert atmosphere, and the coal with particle size ranging from 100 to 200 mesh was sieved for testing. There proximate analyses are given in Table 1.

2.2. Test system

As described in the Introduction part, this study aims to determine the kinetics characteristics of coal low-temperature oxidation based on its thermal characteristics. DSC is a powerful experimental technique to determine the thermal properties of the test sample through testing the difference of the power that inputted to the test sample and the reference material with the increasing temperature (Spink, 2008), so in this study, DSC was adopted, and the DSC instrument used here was the C80 micro-calorimeter produced by SETARAM.

The whole test system was designed as that shown in Fig. 1, mainly including C80 test device, mass-flow gas meter, gas preparation unit, gas cylinders and data processor. C80 microcalorimeter was chosen as the DSC instrument mainly as its high precision of measurement. The 3D Calvet sensor it used totally surrounds the sample and no matter how small the thermal transformation it can provides a complete picture of the event (Setaram). Therefore, it can accurately test the heat generation during coal low-temperature oxidation, although only a small amount of heat is generated, especially in an atmosphere of low oxygen concentration. Meanwhile, the gas circulation vessel (a kind of sample container it provides), with an inlet and an outlet in order to introduce a flow of gas on the sample, allows us to simulate coal oxidation in atmospheres of different oxygen concentration. Mass-flow gas meter could precisely control the flow of gas that pass into the sample chamber and the reference chamber. By using pure oxygen and pure nitrogen, the gas preparation unit can produce gas of different oxygen concentration.

2.3. Test of heat generation of coal low-temperature oxidation in oxygen-depleted air

Testing process mainly included the drying of coal sample, connection of gas pipeline, setting of temperature control program, data collection and the cleaning work at the end of the test.

First, the coal sample was dried in a vacuum oven at 40 °C for 24 h to eliminate the influence of external moisture, and 1000 mg of the dried sample was transferred to the gas circulation vessel. Then, the gas pipeline was connected. Two pipes were connected from the oxygen cylinder and nitrogen cylinder to A and B intake passage of the gas preparation unit respectively, and the configured gas of a certain oxygen concentration was directed to two gas mass

Table 1
Proximate analyses of the coal samples.

Coal samples	Proximate analysis, wt% (air-dry basis)			
	Moisture	ash	VM	FC
Jet coal	3.80	26.31	29.42	40.47
Meagre coal	1.60	11.89	11.33	75.18