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Research article

Assessment of spontaneous combustion status of coal based on relationships between oxygen consumption and gaseous product emissions

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ABSTRACT

It is imperative to have an in-depth understanding of the relationship between oxygen consumption and gaseous product emissions during coal self-heating not only for preventing fires in the coal industry but also for reducing emissions of hazardous gases. Two typical coal samples with high tendency of spontaneous combustion were heated to 230 °C with a programmed temperature of 1.0 K min^{-1} using a pilot-scale test apparatus. The trends for oxygen consumption and gaseous product emissions were obtained from continuous measurements of the gas samples at the reactor outlet via a gas chromatography equipped with a Flame Photometric Detector. Based on the rates of oxygen consumption and gaseous product emissions, the relationship between oxygen consumption and each gaseous product emission was studied. Multi-linear regressions of the oxygen consumption and gaseous product emissions were performed. The coefficients for the emission rates of CO_2, CO and H_2 in the regression equations of the two coals with three different particle sizes are found to be all positive, while the coefficients for the CH_4 emission rates are all negative. These results indicate that the oxygen consumption of coal facilitates the release of CO_2, CO and H_2, while the increase of oxygen consumption rate may not enhance the release of hydrocarbon gas. Based on the results, an evaluation index defined as the ratio of oxygen consumption rate to the gaseous product emission rate is proposed to assess the state of coal spontaneous combustion.

1. Introduction

The spontaneous combustion of coal caused by low-temperature oxidation is a key safety issue in the mining, storage, and transportation of coal, which has restricted and continues to severely restrict the development of the coal industry [1–3]. Along with safety problems, oxidation of coal also significantly affects the molecular structure, elemental composition of coal, and other properties of coal, thereby leading to the deterioration of its technological properties such as carbonization and liquefaction [4,5]. Coal spontaneous combustion also can cause significant environmental problems such as emissions of greenhouse gas and hazardous trace elements [6–8].

Many methods have been proposed to make early detection of the state of coal self-heating in underground coal mines. These methods include monitoring of index gases [9,10], temperature measurements [11–13], numerical modeling [14,15] and radon method [16]. Index gases are a cost-efficient and effective approach to detect coal fires. Analysis of index gases plays a key role for monitoring the status of a suspected heating and predicting the ignition of fire zone. The gaseous products generated from coal self-heating mainly include CO_2, CO, CH_4, H_2, C_2H_4 and other hydrocarbons. Not all these gaseous products released from the low-temperature oxidation of coal are suitable for use as the index gases in coal mines. For examples, CH_4 and CO_2 often occur in coal seams and are therefore not suitable as a single index gas [17]. CO, H_2 and C_2H_4 are commonly used in coal mines as indicators to predict the status of a suspected heating. One of the major concerns with CO as an index gas is that the detection of CO in coal mines does not conclusively indicate a heating event because it can be produced at ambient mine temperature in coal mines [16–19]. Additionally, accurate measurement of O_2 deficiency can also be used to identify the spontaneous combustion status of coal, although there are many problems associated with accurately determining the true amount of oxygen used for use in ratios incorporating oxygen deficiencies [20]. Based on these index gases, several indices, such as production of CO and CO_2 [21], consumption of O_2 [22,23], CO/CO_2 ratio [19], Willet's ratio [24], C/H ratio [25] and Graham's ratio [26] were proposed to

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predict the status of coal self-heating. However, to date no individual index is capable of giving a precise and definite picture of the status of coal heating. These index gases vary case to case depending on the extent and condition of coal fires. Selected index gases have their own limitations. Hence, multiple index gases can help to interpret the extent and development of coal fires more reliably [2,17,19].

Though the developments in the application of index gases over the last few decades have contributed significantly to the management and mitigation of spontaneous combustion risks in coal mines, coal spontaneous combustion incidents still occur which suggest that there is an ongoing need to remain vigilant to this hazard. One of the important reasons is inappropriate and irrational use of index gases. A good understanding of intrinsic relationship between different index gases is critical for timely and accurately predicting the status of spontaneous combustion. Theoretically, the index gas starts to occur when a heating reaches a critical temperature. However, due to the dilution of ventilation and detection limits of the sensors used for gas analysis in underground coal mines, the index gases may not be detectable until the heating has developed to an advanced stage, missing out an optimum time for mine operators to intervene and control the hazard. A robust relationship between index gases and characteristic temperatures of spontaneous combustion has not yet been established.

Before a useful application can be developed, a better understanding of intrinsic relationship between different index gases is required. This would involve further investigation into establishing a robust predicting system for spontaneous combustion. Nevertheless, the intrinsic relationship between different index gases is far from clear, and more research is needed to establish a synergetic prediction system with multiple index gases. In this paper, two typical coal samples with high tendency of spontaneous combustion were heated to 230 °C with a programmed temperature of 1.0 K min^{-1} using a pilot-scale test apparatus. The trend for oxygen consumption and gaseous product emissions was obtained from continuous measurements of the gas samples at the reactor outlet via a gas chromatography equipped with a Flame Photometric Detector. Based on the rates of oxygen consumption and gaseous product emissions, the relationships between oxygen consumption and gaseous product emissions were explored.

2. Experimental

2.1. Coal samples

Two types of coals with high tendency of spontaneous combustion were used in this study. The Shendong coal (denoted as SD), a typical sub-bituminous coal, was obtained from the Chinese Shendong coal mine in the Inner Mongolia. And the Zaozhuang coal (denoted as ZZ), a typical bituminous coal was obtained from the Chinese Zaozhuang coal mine in the south of Shandong province. The two coal mines had badly suffered from coal fires during the last few years [2]. These coal samples were freshly obtained from the recently worked face of the mines, using a chain saw, after first removing a coal layer of approximately 25 cm thickness to avoid the possibility of pre-oxidation. The coal core selected for testing originated from one borehole, thus representing an isorank suite of samples. All samples were first wrapped in a sealed plastic cling wrap, which was completely filled with nitrogen. Next, the cores were transported to the laboratory in an ice-filled insulated container. The cores were then stored in a freezer until required for testing. Before each of the tests, the core was firstly crushed into three different particle ranges: 5.5–7.5 mm, 2.5–5.5 mm, and 0.8–2.5 mm. The samples were then dried at 25 °C in an evacuated desiccator for 24 h. The proximate and ultimate analyses of the coal samples used in this study are presented in Table 1.

2.2. Spontaneous combustion tests

The schematic diagram of experimental apparatus is shown in Fig. 1. The experimental system consists of a programmable oven, a stainless steel reactor, a gas flow system and a multi-channel chart recorder for continuously monitoring of the thermocouple outputs. It can operate in an isothermal or a temperature-controlled style. The reactor body is made of a cylindrical stainless steel tube (i.d. 120 mm, height 800 mm). A pair of porous discs with pore openings of 60 μm diameter, is formed a fixed-bed to charge the coal sample and distribute the incoming gas uniformly over the coal bed. A 0.5-mm-diameter type-K thermocouples are placed at the center of coal. A gas inlet and a gas outlet are located at the bottom and the top of the reactor, respectively.

Table 2 lists the physical parameters of SD and ZZ coals packed in the reactor. Both of the two coals are provided with three different particle ranges: 5.5–7.5 mm, 2.5–5.5 mm, and 0.8–2.5 mm. The data in the tables indicate that the porosity of the coal sample declines with decreasing grain diameter, suggesting that smaller grain diameter leads to closer arrangement of coal particles, smaller space among particles, and lower coal height. Compared to SD coal, ZZ coal with the same grain diameter has a smaller porosity, and the filling height and volume of the coal are relatively small, but the overall amplitude of reduction is relatively low. But as a whole, the differences in features of physical parameters can be regarded insignificant between the two types of coals.

Before each of the tests, the test system was checked of its gas tightness to make sure the system is gas leakage-free. About 1.0 kg coal sample was packed in the reactor and 2–3 mm asbestos was laid on top of the coal samples in order to prevent the gas tube from jamming. The program-controlled temperature enclosure was set to run at a constant temperature of 25 °C. And then air at a flow rate of 2000 cm^3/min (STP) was flowed into the reactor. The program-controlled temperature enclosure was set to run at a programmed temperature of 1.0 °C min^{-1}. During this process, the temperature was monitored continuously and the gas samples at the reactor outlet were analyzed at every 10 °C increase. The exhaust gas exits the container and travels through a water trap before passing through gas chromatography. The reproducibility of the experimental data, checked with some replicate runs, was found to be good.

2.3. Oxidative kinetics parameters

Under the test conditions, the air supply rate in loose coal is constant, and the air flows vertically along the z-axis homogeneously. According to the mass transfer theory, the one-dimensional steady-state equilibrium equation of the concentration of oxygen in coal can be described as follows [27]:

$$Sdx \cdot R_{O_2} = -\mathrm{Q} \cdot dC \tag{1}$$

where, S is the cross-sectional area of the coal tank, m^2; x is the height of coal in the coal tank, m; R_{O2} is the oxygen consumption rate, m^3/(s·m^3); Q is the gas flow rate, m^3/s; C is the oxygen concentration at a special point x; dC is the decreased oxygen concentration when gas flows through the coal sample with the thickness dx in the tank, %.

The oxygen consumption rate of coal at special point x is directly proportional to its oxygen concentration, which can be expressed by Eq. (2):

$$R_{O_2} = \frac{C}{C_o} R_o \tag{2}$$

where, C_o is the standard oxygen concentration of 21%; R_o is the oxygen consumption rate of coal at standard oxygen concentration, mol/(s·m^3); R_{O2} represent the oxygen consumption rate at the oxygen concentration of C.

Substituting Eq. (2) into Eq. (1) leads to Eq. (3),

$$S \cdot dx \cdot \frac{C}{C_o} R_o = -Q \cdot dC \tag{3}$$

Eq. (3) can be re-organized as Eq. (4):