



Full Length Article

Effects of the evolutions of coal properties during nitrogen and MTE drying processes on the spontaneous combustion behavior of Zhaotong lignite

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ABSTRACT

Compared to nitrogen drying process, the mechanical thermal expression (MTE) drying process, a typical non-evaporation drying process with great potential in large-scale application, results in significantly different changes in characteristics of lignite such as pore structures, mineral matters contents and particle size. In this study, the effects of the evolutions of coal properties during nitrogen and MTE drying process on the spontaneous combustion behavior of Zhaotong lignite were systematically investigated. The samples were treated by nitrogen and MTE drying process to desired residual moisture contents, and the changes in pore structures, mineral matters and particle size of samples were analyzed. The test of spontaneous combustion behavior of the sample was carried out by wire basket method. It was found that the removal of water from lignite resulted in the decrease of crossing point temperature (CPT) indicating the promotion of spontaneous combustion susceptibility. The stabilization of pore structures by MTE process which inhibited the transform of macropores into smaller pores, resulted in a lower CPT compared to nitrogen dried samples with the same residual moisture content. The removal of mineral matters from lignite by MTE process also contributed to the decrease of CPT. The effect of particle size in a wide range (from < 0.2 mm to 10–20 mm) on CPT was also studied. The CPT increased with the enlargement of particle size above the critical diameter, due to the less exposition of internal surface area and inhibited the oxygen diffusion to internal particle surface which inhibited the oxidation reactions. The inhibition on the spontaneous combustion of dried lignite by the MTE process was deprived from the large size of its products.

1. Introduction

In China, more than 60% of primary energy is generated from coal in recent years, and the demand of energy has continuously been increasing with the development of economy [1]. As the coal consumption grows, lignite, with as many as 130 billion tons deposit in China, attracts increasingly more attention due to its advantages over black coal, such as low mining cost, high reactivity, high amount of volatiles, and low pollution-forming impurities including sulfur, nitrogen, and heavy metals [2–5]. At the same time, because of the high moisture content in lignite, different methods have been carried out for lignite drying prior to transportation and utilization processes to enhance its competitiveness in energy market [6–8]. However, dried lignite is highly reactive toward oxygen at ambient temperature resulting in the high spontaneous combustion susceptibility which is consequently associated with serious safety problems during storage and transportation

[9–11]. It is therefore of substantial to understand the spontaneous combustion behavior of lignite affected by different drying methods.

The effects of drying processes on the physico-chemical changes of lignite have been widely studied [1,2,12–21]. The predominant chemical changes of lignite are reported to be the destruction of the oxygen containing functional groups (including carboxyl, hydroxyl, and carbonyl) which are significantly affected by the drying temperature and results in the decrease of coal tendency towards spontaneous combustion [2]. It was reported that the breakdown phenolic groups which started at 150 °C became very significant above 200 °C, and that carbonyl groups and free carboxylic acid groups kept up to 150 °C and decompose thereafter [12]. The decomposition of aliphatic hydrogen O–H and C–O groups was observed at 250 °C [13]. Aromatic ring stretch and aromatic carbon remain relatively unchanged up to 250 °C [14]. During air drying, the carboxyl and carbonyl groups increase significantly with the raise of drying temperature until 200 °C, because

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the further increase of drying temperature leads to the decomposition of oxygen functional groups [22,23]. Rare of obvious destruction of the oxygen containing functional groups under 150 °C was reported. In most of the drying process with drying temperature lower than 150 °C under non-oxidizing atmosphere, the chemical changes of lignite are not one of the main factors affecting the spontaneous combustion behavior compared to the changes in pore structure of lignite.

Due to the gel-like porous structure of lignite, the pore structure of lignite is changed with the removal of water during drying process [2,15]. The changes in pore structures directly affect the energy transport and mass transport, which are the most substantial factors of the chemical kinetics reaction between O₂ and active sites on the coal surface during low temperature oxidation. The mesopores of coal are composed of pore channels which chiefly control the transport of gaseous reactants and products [19]. Kaji et al. [24] concluded that the pores with diameters larger than 10 nm played an important role in oxidation of coals at low temperature. Deevi et al. [16] and Li et al. [17] reported that the irreversible changes of pore structure of lignite during drying was associated mainly with the collapse of macroporosity and transitional porosity. Androutsopoulos et al. [18] observed a considerable particle contraction of Greek lignite within the removal of water under vacuum, which led to the decrease of the macro- and partly meso-pore volume and the markedly increase of the surface area. In the further studies on the effects of drying conditions on the pore evaluation, the results indicated that the mesopore volume and surface area increased with the drying temperature until 200 °C, then decreased with further increase of drying temperature [19]. The similar trend of mesopore evaluation was found in hydrothermal dewatering of Chinese lignite by Zhang et al. [1]. The porosity and pore size distribution of lignite were significantly changed by the mechanical thermal expression (MTE) process, which is a typical non-evaporation dewatering process with great potential in large scale application, due to the dewatering and the applied mechanical pressure [20,21,25,26]. During the MTE process, the pores of lignite are almost completely filled by water. The removal of water means the reduction of pore volume. The collapse of macropores contributes to the reduction of pore volume and the increase of mesopores and micropores [20,21]. The changes of pore structure of lignite are considerably different during drying processes by different methods.

In addition, the particle size distributions of lignite dried by varied methods are quite different, resulting in distinct low temperature oxidation characteristics. It is known that the MTE products are briquettes [27], while the nitrogen dried products are featured with mainly in small particles. The mineral matters in lignite also may act as promoters or inhibitors of spontaneous combustion [28–30]. During the MTE process, the soluble mineral matters (such as aluminosilicate, alkalis and alkaline-earth metal) could be removed with water [20,21], which do not occur in evaporation drying process.

The characteristics of dried lignite are therefore significantly affected by different drying methods. However, a full understanding of the effect of drying process on the spontaneous combustion behavior has not been achieved. The effect of air drying on the spontaneous combustion characteristics of Turkish lignite has been reported by Kadioğlu et al. [9]. Zhao et al. [10,31] compared the spontaneous combustion behavior of Indonesian lignite prepared by nitrogen drying and vacuum. The moisture content and particle size are recognized to be important factors which influence the spontaneous combustion susceptibility of lignite. The decrease of moisture content and particle size reduced the liability of lignite to spontaneous combustion [9,10,31,32]. But there is still lack of information about the effect of particle size on lignite spontaneous combustion at the range of diameter larger than 1 mm, while the size of MTE product could exceed 100 mm [27]. The different changes of pore structures during nitrogen and vacuum drying process also exert considerable influence on the spontaneous combustion susceptibility of pre-dried lignite [10]. But due to the further drying required by pore structure test, the sample for

spontaneous combustion test is not the same sample for pore structure test, in strictly. Further research should be carried out to investigate the effect of drying processes on the spontaneous combustion behavior of lignite.

Zhaotong lignite, whose structure and upgrading has been widely studied as a typical lignite in China [33–35], is suitable to be dried by MTE process in that its moisture content is higher than 50% [4]. As mentioned above, the MTE process would result in the significantly different changes of pore structures, mineral matters content and particle size of lignite compared to evaporation drying process. However, a thorough understanding of the effects of these factors on the spontaneous combustion behavior of lignite has not been achieved. In this study, the lignite obtained from Zhaotong (Yunan, China) was separately dried by nitrogen, a typical evaporation drying process, and MTE drying process to desired residual moisture contents. The spontaneous combustion behavior of dried lignite was tested by wire basket method. The effects of the changes in pore structures, mineral matters content and particle size of lignite during the drying processes on its spontaneous combustion behavior were discussed.

2. Experimental

2.1. Coal preparation and analysis

The lignite used in this study was obtained from Zhaotong, Yunnan, China. The characteristic of this raw coal sample was given in Table 1. The proximate analysis was carried out following the ISO 11722, ISO 1171 and ISO 562 methods. The ultimate analysis was carried out following the ISO 625, ISO 333 and ISO 334 methods.

The raw lignite was crushed and sieved into different sizes (< 0.2 mm, 0.2–0.5 mm, 0.5–1 mm and 1–2 mm, respectively), then dried under nitrogen at 105 °C. The MTE drying process was carried out as literature [27]. The briquette produced by MTE process (150 mm in diameter in this study) was crushed into different sizes. To eliminate the influence of chemical changes caused by different drying temperature on the spontaneous combustion behavior of lignite, all the drying experiments were carried out at 105 °C. The samples with different residual moisture contents were obtained by controlled drying time. The samples were named by drying method and residual moisture content. Such as, Nitrogen-20% and MTE-20% represented samples with 20% residual moisture content prepared by nitrogen drying and MTE drying, respectively.

2.2. Pore analysis

A BELSORP-max ver 2.1 gas adsorption instrument (Bel) was applied to analyze the micropores and mesopores structures of the samples by N₂ adsorption method. The samples were degassed under vacuum at 105 °C for 4 h and then placed in the instrument for test. The Brunauer-Emmett-Teller (BET) model was used to characterize the surface area. The Barrett-Joyner-Halenda (BJH) model was employed to calculate the pore volume between 1.2 nm and 50 nm which includes mesopores and part of micropores.

Table 1
Proximate and ultimate analyses of the raw coal sample.

Samples	Proximate analysis (wt%)				Ultimate analysis (wt%)				
	M _{ar}	A _d	V _{daf}	FC _{daf} [*]	C _{daf}	H _{daf}	N _{daf}	S _{daf}	O _{daf} [*]
Zhaotong	56.12	19.36	59.17	40.83	65.78	4.06	1.97	1.85	26.34

ar: as receive basis, d: dry basis, daf: dry ash free basis, M: moisture, A: ash, V: volatile matter, FC: fixed carbon.

*By difference.

The errors are: M_{ar}, ± 0.3%; A_d, ± 0.1%; V_{daf}, ± 0.3%; FC_{daf}, ± 0.5%; C_{daf}, ± 0.2%; H_{daf}, ± 0.06%; N_{daf}, ± 0.04%; S_{daf}, ± 0.03%; O_{daf}, ± 0.3%.

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