



# Effects of drying method on self-heating behavior of lignite during low-temperature oxidation



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## ABSTRACT

Pore structure changes during pre-drying of lignite affect its low-temperature oxidation and increase the susceptibility to spontaneous combustion. In this study, the effects of drying methods (i.e., vacuum drying and N<sub>2</sub> drying) on self-heating of Indonesian lignite during oxidation were investigated using a dual fixed-bed quartz reactor. The variation of coal temperatures was recorded and the release of CO<sub>2</sub> and CO was measured by a gas chromatography. The pore volume and surface area of dried samples were measured using Brunauer-Emmett-Teller (BET) method. Mesopores in lignite initially increased and collapsed with further increasing drying intensity during drying in N<sub>2</sub>, resulting in a rapid self-heating rate of lignite within a critical moisture content range of 6–13%. However, vacuum drying caused a gradual increase in mesopores, which lead to a monotonic increase in self-heating rate with decreasing residue moisture content in lignite. The experimental results indicated that the production rates of both CO<sub>2</sub> and CO during oxidation of raw lignite increased with reducing particle size and increasing gas flow rate, but decreased at lower moisture contents. Typically, the variation of production rates of both CO<sub>2</sub> and CO as a function of particle size and gas flow rate followed a similar trend to that of raw lignite when the lignite was completely dried by the vacuum drying method. The impacts of lignite particle size and gas flow rate on the yields of CO<sub>2</sub> and CO was limited due to less diffusion of O<sub>2</sub> into small pores, suggesting that the oxidation reaction between lignite and oxygen has been shifted from diffusion controlled to kinetic controlled reactions.

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## 1. Introduction

The high moisture content (25–65%) of lignite results in lower calorific value, higher fuel consumption, higher power consumption, lower efficiency and higher transportation cost [1–3]. The drying of lignite prior to use or transportation is expected to increase its competitiveness in energy market [4]. However, lignite especially in dry state [3,5,6], is highly reactive toward oxygen at ambient temperature and is highly susceptible to spontaneous combustion as a result of low-temperature oxidation [2].

The physical changes of lignite during drying have been extensively reported in the literature [1]. The porous structure of lignite is comprised of micropores ( $\chi < 2$  nm,  $\chi$  = pore diameter), mesopores (2 nm  $< \chi < 50$  nm), and macropores (0.05  $\mu\text{m} < \chi < 60$   $\mu\text{m}$ ) [7–9]. The porous structure of lignite is gel-like which shrinks in response to water loss [1,10–13]. The contraction of coal particles due to drying

alters the original pore structure of the lignite [11,14]. Li et al. [14] measured the moisture desorption and re-adsorption isotherms of Indonesian lignite from bed moist state, showing that irreversible structure changes took place in coal during drying. The irreversible shrinkage of lignite was associated mainly with collapse of macroporosity and transitional porosity [10]. Hulston et al. [15] observed the decrease in total pore volume of lignite with increasing drying temperature, which was attributed to the progressive collapse of macropores and a slight increase in mesopore volume. Androustopoulos and Linaros [12] studied the effect of drying upon the macro- and partly meso-pore structure of Greek lignite. They observed that isothermal drying under vacuum caused a considerable particle contraction (by  $\sim 1/3$  of its original size), a minor decrease in the macro- and partly meso-pore volume and a marked increase in the surface area, particularly in the range of high weight losses (35–40%). They also reported the formation of pores in the size range of 7.5–150 nm at the expense of pores in the 150–1000 nm range during drying. Salmas et al. [16] reported the evolution of mesopore structure during thermal drying of Greek lignite despite the severe ( $\sim 50\%$ ) particle size contraction. Mesopore volume and surface area increased as the drying temperature was increased to 200 °C while further drying up to 250 °C caused their decrease.

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**Table 1**  
Proximate and ultimate analysis of the coal used in this study.

Proximate analysis (ad)		Ultimate analysis (daf)	
Moisture (%)	24.06	C (%)	71.51
Ash (%)	2.170	H (%)	4.970
Fixed carbon (%)	36.53	N (%)	1.028
Volatile matter (%)	37.07	S (%)	0.242
Total sulfur (%)	0.170	O (%)	22.25

ad = air-dried; daf = dry ash free.

The self-heating of lignite during oxidation is governed by  $O_2$  diffusion toward the surface of pore walls and chemical kinetics reaction between  $O_2$  and active sites on the surface, depending on the intrinsic reactivity of the coal and the capacity of coal pores to transport oxygen [9,17–21]. There is no doubt that the changes in pore structure characteristics during pre-drying directly affect the mass transport and chemical reaction mechanism involved in oxidation process [1]. Kaji et al. [19] concluded that small pores (<10 nm) did not contribute to the oxidation reactions at low temperature. Coal mesopore structure is composed of pore channels that mainly control the transport of gaseous reactants and products [16]. Bouwman and Freriks [22] proposed two models for coal oxidation to explain the effect of particle size, i.e. macropore oxidation and micropore oxidation. Oxidation of large particles follows the macropore oxidation model in which transport of  $O_2$  is the rate-limiting step, while micropore oxidation occurs in small particles and is not limited by oxygen diffusion.

The pore structure of lignite dried by different drying methods varies considerably, resulting in different low-temperature oxidation characteristics. However, a full understanding of the exact mechanism of low-temperature oxidation of the dried lignite has not yet achieved. In this study, an Indonesian lignite was subjected to vacuum drying at 45 °C and  $N_2$  drying at 105 °C. Lignite samples with different moisture contents were prepared at different drying residence time. The self-heating of the dried samples during low-temperature oxidation was investigated using a dual fixed-bed quartz reactor. The pore structure of coal samples and the formation of gaseous products during oxidation were evaluated and discussed in detail. The main objective of this study is to investigate the effect of pore structure changes during pre-drying on low-temperature oxidation and self-heating of lignite.

## 2. Experimental section

### 2.1. Sample preparation and analysis

An Indonesian lignite was used in this study and its proximate and ultimate analyses are given in Table 1. The raw Indonesian lignite was crushed and sieved under nitrogen into four different size fractions (125–150, 220–300, 355–450 and 500–600  $\mu\text{m}$ , respectively). The

samples were then placed and labeled in sealed containers and stored in a fridge. The coal samples were dried at 45 °C under vacuum and at 105 °C under nitrogen. The samples with different moisture contents were obtained by controlling drying time either under vacuum or in  $N_2$ .

The BET surface area and pore volume of all samples used in this study were measured by a specific surface area and pore size analyzer (V-sorb 4800 P). Nitrogen was used as the adsorption gas at 77.3 K. The samples were first vacuum dried at 30 °C and then degassed in the adsorption system at 200 °C at a final pressure of  $1.33 \times 10^{-4}$  Pa. The Barrett–Joyner–Halenda (BJH) [23] model was used to determine the pore size distributions from the desorption isotherm. The  $N_2$  adsorption method can characterize the pores with diameter between 1.7 nm and 300 nm, which mainly includes mesopores and part of macropores. This method cannot adequately measure the total pore volume and surface area of coal, but is valuable in characterizing larger pores relevant to transport of oxygen during oxidation [9].

### 2.2. Oxidation experiments

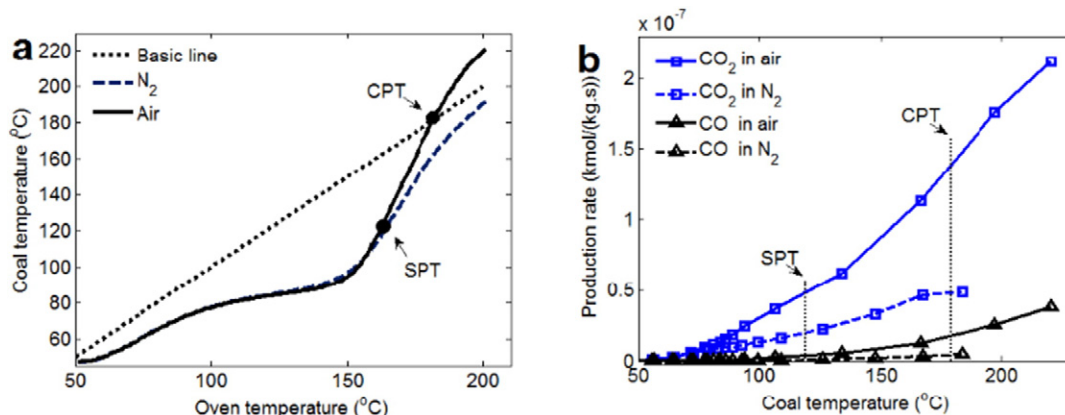
The oxidation experiments were carried out in two vertical fixed-bed tubular quartz reactors ( $\varphi = 2.0$  cm) heated in an programmable electric furnace (SY60, Shenyang General Furnace Manufacturing Co., Ltd., China). Details of the experimental apparatus have been reported in our previous study [21]. A sample of 28 g was placed into two reactors. Temperatures of the oven and two coal-beds under nitrogen were monitored by three thermocouples. Once these three temperatures became consistent, the low temperature oxidation experiment was started by switching nitrogen gas to air at the same flow rate. The furnace was heated from 50 °C to 200 °C at the rate of 1 °C/min. Coal-bed temperatures during experiment under both air and nitrogen were recorded in data acquisition system (Advantech USB-4718, Taiwan). The concentrations of  $CO_2$  and CO at the outlet of reactor were measured using a gas chromatograph (G5, Beijing Puxi General Instrument Co., Ltd., China) equipped with a methane conversion oven and a flame ionization detector (FID). The production rates of carbon oxides were calculated using the following equations [18,21,24–26]:

$$R_{CO_2} = C_{CO_2} \cdot V_{gas} / W \quad (2)$$

$$R_{CO} = C_{CO} \cdot V_{gas} / W \quad (3)$$

where,  $C_{CO_2}$  and  $C_{CO}$  are carbon dioxide and carbon monoxide concentrations at the reactor exit, respectively.  $V_{gas}$  is the flow rate of the carrier gas and  $W$  is the dry mass of a coal sample.

An example of temperature profile of the coal beds and the yield of the gaseous products is shown in Fig. 1. The separation point temperature (SPT) was defined as the point at which the coal temperature in air was initially higher than that in nitrogen and was considered as



**Fig. 1.** (a) Changes in temperatures of coal-bed and (b) production rates of  $CO_2$  and CO for raw lignite during oxidation.

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