Applied Thermal Engineering 143 (2018) 542–549

Contents lists available at ScienceDirect

Applied Thermal Engineering

journal homepage: www.elsevier.com/locate/apthermeng

Research Paper

Thermal properties of coals with different metamorphic levels in air atmosphere

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HIGHLIGHTS

- Variation of crystallinity of coal with different metamorphic levels was explored.
- Effect of coal crystallinity on its thermal diffusivity was investigated.
- Thermal properties of coal with different metamorphic levels in air were researched.
- Correlation among thermal property parameters was achieved.

ARTICLE INFO

Keywords:
Heat transfer
Crystallinity
Thermal diffusivity
Specific heat
Thermal conductivity

ABSTRACT

Thermal properties of coal are crucial in the heat-transfer process of its spontaneous combustion. Thermophysical properties of four metamorphic-grade coal samples were investigated in air using a laser-flash apparatus. Their crystallinities were analyzed by X-ray diffractometry. The results indicated that the trend in thermal diffusivity was opposite to that of crystallinity. As the temperature increased, thermal diffusivity first decreased and then increased; specific heat first increased and then decreased. With the exception of the meager lean coal sample, the trend of thermal conductivity as a function of temperature agreed with that of specific heat. As the metamorphic grade of the samples increased, the thermal diffusivity minimum and specific heat maximum shifted toward higher temperatures; in contrast, the minimum thermal conductivity shifted toward lower temperatures. From these trends, it was possible to recognize different temperature ranges according to how different properties (thermal conductivity, thermal diffusivity, specific heat, and metamorphism grade) influenced each other.

1. Introduction

China is among the major coal-producing countries of the world [1,2]. Coal constitutes a large proportion of the energy consumption in China and, although production has declined in recent years, coal remains the main energy source [3]. Spontaneous coal combustion is one of the main threats to mine safety and the environment [4,5] and is of growing concern [6,7]. When coal is oxidized and undergoes an exothermic reaction, heat transfer is engendered. During the heat-transfer process of spontaneous coal combustion, thermal properties play a vital role.

Thermophysical parameters are intrinsic factors determining the heat-transfer ability of a material; therefore, the thermal properties of coal–rock masses have drawn the attention of researchers. Herrin et al. [8] explored the thermal conductivity of different coal samples at room temperature and analyzed the effects of moisture, ash, volatiles, and substrates. Seipold [9,10] established a calculation model of thermal conductivity and thermal diffusivity by studying the effect of temperature on the thermal properties of a crystalline rock sample. Pertermann et al. [11,12] analyzed the effect of crystal direction and elemental composition of a rock sample on its thermal properties by measuring the thermal diffusivity of olivine from room temperature (ca. 20 °C) to 1500 °C. Abdulagatova et al. [13,14] discussed the effect of pore size and minerals on the thermal properties of coal rocks, and

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https://doi.org/10.1016/j.applthermaleng.2018.07.117
Received 11 May 2018; Received in revised form 23 July 2018; Accepted 23 July 2018
Available online 24 July 2018
1359-4311/

analyzed the variation of thermal conductivity of sandstone, limestone, and other rocks with pressure and temperature. Vosteen et al. [15] studied the variations of thermal conductivity, specific heat, and thermal diffusivity of crystalline, sedimentary, and other rocks with temperature, and proposed a corresponding calculation model. Varma et al. [16,17] investigated the thermal conductivity of coal and rock samples obtained from the Ruqigou coalfield by conducting field sampling tests and measuring the thermophysical properties of various rocks using the flash method. Hofmeister [18–20] measured the effect of temperature on the thermal diffusivity of garnet and clinopyroxene using a laser-flash apparatus and proposed a calculation model. Merriman et al. [21] studied the heat transfer properties of major Archean rock types under high temperature, and revealed that the surface geothermal contributes less to the heat production of the crustal by using numerical models. Miao et al. [22] also analyzed variations of thermal conductivity and thermal diffusivity of different rock types with temperature using this apparatus.

Spontaneous coal combustion occurs because coal absorbs oxygen at room temperature, thus resulting in low-temperature oxidation and the release of heat and primary oxidation products. Owing to poor heat dissipation, the low-temperature oxidation process is accelerated by the accumulation of heat and increase in temperature, which eventually leads to spontaneous combustion. Coal oxidation and heat storage are among the necessary conditions for spontaneous combustion. Heat transfer is involved in coal oxidation. Coal samples with different metamorphic grades have different heat-transfer capacities, which are related to their thermal properties. Therefore, the objectives of this study were to (1) investigate the crystallinity levels of coal samples with different metamorphic grades; (2) explore the variations of thermal conductivity, thermal diffusivity, and specific heat with temperature in an air atmosphere; and (3) analyze the correlations between these thermophysical parameters. The results of the study provide insight into the heat conduction of coal of different metamorphic grades during spontaneous coal combustion.

2. Experimental methods

2.1. Coal samples

As described in Table 1, four types of coal samples with different metamorphic grades—lignite, long-flame coal, gas coal, and meager lean coal—were selected. The A and B samples were collected from Inner Mongolia, China; the C and D samples were obtained from Shandong and Shanxi Provinces, China, respectively. The results obtained from the proximate analysis of the four samples are listed in Table 1.

Table 1

2.2. Experimental conditions

Before each experiment, the sample was pulverized to a particle size of no greater than 0.074 mm. The powdered sample was pressed into tablets using a YP-2 tablet-pressing machine. The thickness and diameter of each tablet were approximately 1.0 mm and 12.7 mm, respectively. Each tablet was placed into the instrument and air was supplied into a furnace at a rate 1.67 $10^{-6}\,m^3/s$. To alleviate thermal inertia, the sample was heated from 30 to 300 °C at a heating rate of 1 °C/min. A linear baseline was selected and the Cowan and pulse correction modes were set. To ensure the highest possible accuracy of the specific heat measurement, the sample and reference were of the same surface shape, surface smoothness, and area, and used the same sample tray and light shield to ensure the same absorption and infrared emission area. Simultaneous graphite coating of the sample and reference ensured that their surface absorption rates and emissivities were strictly consistent, avoiding the influence of different colored samples on the light absorption and radiation ability. Each tablet was measured three times after each 30 °C increment. The thermophysical parameters for each collection point were obtained by calculating the average of three measurements.

Table 1
Proximate analyses of four coal samples—Coal A to Coal D (air-dried basis, mass%).

Degree of metamorphism	Specimen	Moisture	Ash	Volatile	Fixed carbon
Lignite	Coal A	15.14	6.42	52.61	25.83
Long flame coal	Coal B	9.49	9.19	45.26	36.06
Gas coal	Coal C	2.45	8.85	33.42	55.28
Meager-lean coal	Coal D	0.69	7.83	13.59	77.89

Note: the degree of metamorphism of coal refers to the degree of change in the physical and chemical properties of coal under the interaction of temperature, pressure and time.

To maintain the original characteristics of the coal samples, they were not treated for ash removal. The prepared samples were loaded on an aluminum frame for X-ray diffraction (XRD) analysis. Experiments were conducted using copper target radiation and continuous scanning under a pipe pressure of 40 kV and pipe flow of 30 mA. The coal samples were scanned from 10° to 80° at 6°/min.

2.3. Instrumental methods

2.3.1. Laser-flash apparatus

The thermophysical parameters of the four coal samples were examined using a laser-flash apparatus (LFA457, Netzsch GmbH, Germany), which primarily included a temperature detector, high-temperature furnace, laser heating system, and data acquisition system. A laser source or flash xenon lamp emits a light beam, which is evenly irradiated on the lower surface of the samples. The temperature increases instantaneously as the surface absorbs the light energy. The hot end propagates the energy upward in a one-dimensional heat conduction mode. The upper surface center temperature of the samples was continuously measured by an infrared detector. The operating principle of LFA457 was previously described [23,24].

2.3.2. X-ray diffractometer

To determine their crystallinity, the four coal samples were tested by X-ray diffractometry (XRD-7000, Shimadzu, Japan). Before each measurement, the circulating condensate and panel switches were turned on to control a certain circulating water temperature. After preheating the instrument, a tablet with a flat surface was placed in the instrument. The sample was scanned by setting the sample parameters and test conditions, and turning on the X-ray tube voltage.

3. Results and discussion

3.1. Crystallinity and thermal diffusivity

Fig. 1 presents the fitted XRD patterns of the four coal samples. The areas of the amorphous and crystal peaks of the samples were estimated by fitting. The crystallinity of each sample was determined using Eq. (1) and the values are presented in Table 2:

$$X_c = (I_a/I_t) \times 100\% \tag{1}$$

where X_c is the absolute crystallinity (%), I_a is the area of the crystalline peak, and I_t is the total area of all peaks [25].

With an increase in the metamorphic grade of the sample, the integral strengths of the crystalline and amorphous peaks gradually increased, except for Coal A. Eq. (1) revealed that the degree of crystallinity is not only related to the crystal peak area, but also to the amorphous peak area. Because the integral intensities of the amorphous peaks increased faster than those of the crystalline peaks, the crystallinity variations were not consistent with the variation of the degree of