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Volatilization of mercury in coal during conventional and microwave drying and its potential guidance for environmental protection

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ABSTRACT

During the thermal drying process of coal, the sufficient attention has been paid to the changes in the properties of coal before and after the drying process. However, the volatility characteristics of harmful trace elements in coal during the drying process are always neglected to date, whereas the volatilization of harmful trace elements not only has significant effects on the surrounding atmosphere environment but also influence the following utilization of dried coal. In this study, the volatility characteristics of both the moisture and mercury in the coal sample during conventional (electric blast oven) and microwave (microwave oven) drying processes were investigated. In addition, the changes in the surface properties of coal before and after the drying process were studied using X-ray photoelectron spectroscopy (XPS) analysis. The results show that the moisture content of coal decreases in non-line format (decreases slowly at the first and then quickly according to the drying time) in the conventional drying process, while it decreases in line format in the microwave drying. The microwave drying process needs much shorter time required to obtain the dried coal of a certain moisture content than the conventional drying process. Under the same volatilization rate of moisture, the volatilization rate of mercury in the microwave drying process is always greater than that in the conventional drying process. Nearly half or more (49.73% and 59.26%, respectively) of the mercury in the coal slime evaporates into the atmosphere after the drying processes. XPS results show the content of C—C and C—H groups on the coal surface increased, but the C—O content decreased after the drying processes and the microwave drying process has a stronger ability to remove oxygen-containing functional groups than the conventional drying process.

1. Introduction

Coal is still one of the main energy resources in the world (Burmistrz et al., 2016a, 2016b). As one of the main products in the wet coal preparation process, coal slime has the characteristics of fine size composition and high moisture content. Coal slime must be dried in order to increase the calorific value, decrease the transportation and handling costs, improve the combustion efficiency, wide the application directions, and decrease the power plant emissions (Argiz et al., 2018; Pietrzak and Wachowska, 2004). The coal slime drying method can be divided into two categories: mechanical drying and thermal drying. Mechanical drying includes the processes of sieving, centrifugation, and filter press drying. However, the moisture content is still high (approximately 20%) and fails to meet the requirements of many industrial applications using mechanical drying solely. Thermal drying is usually required to further lower the moisture content (Song et al., 2017). Thermal drying can be divided into two categories, one is the conventional drying method represented by hot air drying, and the other is a new type of drying method named microwave drying (Tahmasebi et al., 2011). Generally, coal is a mixture of organic and inorganic materials, therefore coal is easy to be oxidized in air as well as in the acid/alkaline solutions (Grzybek et al., 2004; Pietrzak and Wachowska, 2004). The oxidation degree of coal during the dry process also needs sufficient attention.

In the thermal drying process of coal slime, the concerns are often paid on the dehydration efficiency and energy consumption, but the environmental problems are ignored, such as the pollution

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of trace elements during the drying processes. Trace elements in coal have been extensively studied due to their harmfulness (Masto et al., 2011; SWAINE, 1998). Many papers have shown that many trace elements in coal tend to be enriched in fine coal slime (Pan et al., 2016; Zhou et al., 2016, 2017). Therefore, the release/volatilization law of the harmful trace elements, especially volatile elements (e.g. mercury), to the atmosphere during the thermal drying process of coal slime should be given sufficient attention.

As a common harmful volatile element in coal, mercury has received many attentions as its pollution has become a worldwide environmental problem (Huang et al., 2016; Li et al., 2015; Zhang et al., 2014). However, the volatility of mercury during the thermal drying processes of coal slime has never been reported to date. In the conventional drying processes, the heat transported from the surface of coal to the inner particles. For the microwave drying, the materials absorb the energy according to their own properties (Beary, 1988). The comparison of volatility characteristics of moisture in coal using the conventional and microwave drying has already shown the microwave can achieve a relative quicker and more efficient dewatering rate *vs.* drying time (Tahmasebi et al., 2012b). Therefore, the volatility characteristics of harmful trace elements in coal using the conventional drying process should also differ from that using the microwave drying. This work is attempted to study the effects of the conventional drying process and microwave drying process on the volatility of both moisture and mercury in coal slime. The changes in the properties of coal before and after the drying process were also investigated using X-ray photoelectron spectroscopy (XPS) analysis.

2. Materials and methods

2.1. Coal sample

The coal slime sample was taken from a Coal Preparation Plant located in Ningxia, which is one of the main producing areas of high-Hg coals in China. The coal slime sample was sealed to prevent the natural evaporation of moisture. The particle size of the solid particles in the slime was detected to be −74 μm. The content of moisture, ash, and mercury of the raw coal slime sample are shown in Table 1.

2.2. Experiment setup and test procedure

2.2.1. Coal slime drying

The heating tool of the conventional drying process is an electric blast oven in this study, while that of the microwave drying process is a microwave oven. A porcelain boat containing 3 g of raw coal slime sample was placed in an electric blast oven or in a microwave oven for the selected duration. The electric blast oven was kept constant at 200 °C and the power of the microwave oven was set at 800 W. Three coal samples were dried in parallel for each drying time in each drying process. After the drying, the mass of each parallel coal sample was weighed, and the residual moisture contents of the sample after the drying and the cumulative volatilization proportion of moisture were calculated by means of the following equations (Eqs. (1) and (2)), respectively. The average of the measured parameters of the three parallel dry coal samples is reported in the paper.

$$R_M = \frac{m_{d,t} - m_i \times (100\% - M_{ar})}{m_{d,t}} \times 100\% \quad (1)$$

$$V_M = \frac{m_i - m_{d,t}}{m_i \times M_{ar}} \times 100\% \quad (2)$$

where R_M is the residual moisture content, V_M is the cumulative volatilization proportion of moisture, m_i is the initial sample mass, $m_{d,t}$ is the mass of the dried coal after the drying time of t, and M_{ar} is the moisture content (as received basis).

2.2.2. Determination of mercury content

All samples (including raw coal samples and dried coal samples after different drying times in different drying processes) were allowed to stand in flowing air at room temperature for 48 h until the mass of the samples did not change (that is, air dry state, the moisture content was about 1.85%). Then the mercury content in all of the solid samples was detected by a DMA-80 Direct Mercury Analyzer (Milestone Srl., Italy). The cumulative volatilization proportion of mercury was calculated by means of Eq. (3).

$$V_{Hg} = \frac{C_{Hg,i} - C_{Hg,d,t}}{C_{Hg,i}} \times 100\% \quad (3)$$

where V_{Hg} is the cumulative volatilization proportion of mercury, and $C_{Hg,i}$ and $C_{Hg,d,t}$ (air-dried basis) is the mercury content in initial sample and in the dried sample after the drying time of t, respectively.

2.2.3. XPS analysis

Air-dried coal samples were subjected to XPS detection. The XPS experiments were carried out at room temperature in an ultra-high vacuum (UHV) system with the surface analysis system (ESCALAB 250Xi, America). The data processing (peak fitting) was performed with XPS Peak fit software, using a smart type background subtraction and Gaussian/Lorentzian peak shapes. The binding energies were corrected by setting the C1s hydrocarbon ($-CH_2-CH_2-$ bonds) peak at 284.6 eV.

3. Results and discussion

3.1. Volatilization of moisture and mercury during drying processes

The changes of moisture and mercury contents with the drying time were plotted in Fig. 1. The moisture content was reduced with the drying time. When the moisture content of coal slime was dried to a very low level (~1.5%), the drying time required in the microwave drying process (120 s) was much shorter than that required in the conventional drying process (6 min and 40 s). The mercury content of the coal slime using the microwave drying was generally lower than that using the conventional drying. In the conventional drying process, the decrease rate in moisture was slow first and then faster, while the reduction rate of mercury was extremely low or not significantly changed. However, in the microwave drying process, the moisture content decreased in line format while the mercury content changed little in the initial phase of the drying process (<90 s) and then declined rapidly in the late drying process (90–120 s).

Fig. 2 illustrates the relationship between the volatilization amount of mercury and that of moisture under the two drying processes. The results show that the volatilization amount of

Table 1
The content of moisture, ash, and mercury of the raw coal, wt. %.

M_{ar}	M_{ad}	A_{ad}	V_{daf}	FC_{daf}	Hg_{ad}[a]
21.48	1.85	21.61	10.65	89.35	338.387

M: moisture; A: ash; Hg: mercury; ar: as received basis; ad: air-dried basis; daf: dry ash free.
[a] μg/kg.