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Characteristics of polycyclic aromatic hydrocarbon release during spontaneous combustion of coal and gangue in the same coal seam

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ABSTRACT

Based on the influence of temperature, coal quality, oxygen supply and other factors, the formation characters of PAHs during spontaneous combustion of coal and gangue in the same coal seam were experimentally studied. The results showed that the amount of PAHs released from coal and gangue first increased and then decreased with the rising of coal and gangue spontaneous combustion temperature. And it reached a peak value about 5.76–7.17 times of the minimum value at 400 °C–500 °C. The types of PAHs released during the high temperature stage of spontaneous combustion were up to 9. The release amount of PAHs generally increased with the rising of the volatile content, sulfur content, carbon content, fixed carbon content and calorific value of coal and gangue, but decreased with the rising of ash content. The amount of PAHs produced by spontaneous combustion of raw coal was generally higher than that of gangue, and the difference was more obvious at high-temperature stage. The total toxicity amount of PAHs produced by spontaneous combustion of raw coal was higher than that of gangue, up to 9.93 times. Its law of change with temperature was consistent with the total amount of PAHs, which increased and then decreased with the rising of temperature; and reached the peak at 300 °C–400 °C with a peak value of 6.72–14.60 times of the minimum value.

1. Introduction

At present, the pollution caused by the coal (gangue) spontaneous combustion process and its treatment have got widely concern around the world. In spontaneous combustion of coal and by-product coal gangue, in addition to conventional pollutants and dust like NO_x, CO_2 and SO_2, some special organic pollutants are also released, including olefins, alkanes and aromatics, as well as polycyclic aromatic hydrocarbons (PAHs), which have a serious impact on the environment (Pone et al., 2007; Finkelman, 2004; Zhou and Zhao, 2012). With coal mining, spontaneous combustion of coal gangue has caused many environmental pollution problems, of which PAHs are particularly harmful due to their bioaccumulation and chronic effects of carcinogenic, teratogenic and mutagenic.

The researches on the release of PAHs during the combustion of coal (gangue) mainly focus on two aspects: the PAHs inherent characteristics as well as the general rules of the mechanism and yield of releasing PAHs during oxygen-rich combustion of coal (gangue). Pergal et al. (Pergal et al., 2013) found that the amount of PAHs generated from coal combustion first increased at a temperature of 930 °C–998 °C, but remained unchanged from 973.5 °C. Misz and Fabiańska (Misz-Kenna et al., 2011) put forward the factors that affected the generation of PAHs during combustion of coal (gangue), such as combustion type, combustion temperature, ventilation rate and so on. Master, Callén and Garcia (Master et al., 2000) argued that the combustion temperature and the formation of PAHs were not directly linked. Liu, Wen, Pan, et al. (Liu et al., 2001a, b) found that under the combustion environment of 850 °C–900 °C, PAHs could be chemically converted when they were destroyed by HNO_3 and the like simultaneously. Through different types of bituminous coal combustion experiments, Liu et al. (2010) concluded that the generation amount of PAHs increased and then reduced with the increase of carbon content, and reached the highest point under the carbon content of 80%–85%. Zhao et al. (2000) pointed out that PAHs released by coal combustion reached the highest when the volatile content was around 40%. Yao et al. (2003) concluded that the amount of PAHs produced during coal combustion got the highest when the molar ratio of oxygen to carbon was about 0.1. Liu et al. (Liu et al., 2001a, b; Liu, 1999) believed that the generation amount of PAHs increased with the rising volatiles of coal. The amount decreased under the high ash content and low volatile matter content (Liu et al., 2000a, b), and increased under the low coal heat. The structure of coal itself is also one of the important factors that affect the generation of PAHs. Generally, the amount of PAHs produced by spontaneous combustion of low rank coal is much higher than that of high rank coal (Liu et al.,

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Table 1
The basic parameters of samples selected.

Sample	Proximate Analysis				Elemental Analysis				$Q_{b,ar}$/ J/g
	M_{ad}/%	V_{ad}/%	A_{ad}/%	FC_{ad}/%	N/%	C/%	S/%	H%	
FHM	1.09	10.19	12.70	76.03	0.97	81.63	2.23	2.58	29332.21
FHG	1.11	9.48	85.49	3.93	0.28	74.82	1.56	1.01	1143.06
YCM	1.16	9.68	20.67	68.50	0.68	77.36	0.33	2.12	25248.12
YCG	1.07	8.08	88.80	2.06	0.55	70.36	0.23	1.05	917.70

2000a, b).

Different from general coal combustion, spontaneous combustion of coal (gangue) belongs to the slow oxidation process in oxygen-lean condition. In most cases, its temperature is in the middle and low range. The seepage characteristics of airflow in the accumulated coal (gangue) are also different from the condition of airflow and the mode of oxygen supply in the state of pulverized coal. The above research results provide technical support and basic theory for enriched coal combustion theory and environmental protection, which however can not be used to evaluate the release characteristics of PAHs during spontaneous combustion of coal (gangue), nor to quantify the relevant environmental effects. On this basis, through simulation theory of coal spontaneous combustion and supported by technical equipment, combined with industrial analysis and elemental analysis of coal (gangue), comprehensively considering the impact of temperature, coal quality, oxygen supply and other factors, we studied PAHs generation law during the spontaneous combustion process of raw coal and gangue in the same coal seam. It focused on comparative analysis of PAHs generated characteristics from raw coal and gangue in the same coal seam, in order to provide theoretical support for the effective determination of PAHs generation during spontaneous combustion of coal gangue.

2. Experimental area and samples

Jincheng city is located in the southeastern of Shanxi Province at the junction of Shanxi and Henan. Its most part is within Qinshui Basin, which contains rich coal resources. The Jincheng mining area is mainly located in the southeast of the Qinshui Basin, southwest of the Taihang Mountains, and north of the Qinling Mountains. There are Wangtaipu coal mine, Fenghuangshan coal mine, the Gushuyuan coal mine and other old mines in the mining area, which have been mined for a long time to produce a large number of coal gangue hills. The Taiyuan Formation and Shanxi Formation in the mining area are all coal-bearing formations with overall 13 layers and a total thickness of 13–15 m. The thickness of Taiyuan Formation is 80–100 m with 10 layers of coal,. The thickness of Shanxi Formation is 40–80 m, with mudstone and 1 to 3 layers of coal seams.

The coal deterioration degree and sulfur content are important internal factors that affect the generation of PAHs during spontaneous combustion of coal and gangue. For this reason, the paper selected No. 3 coal from Shanxi Yuecheng coalmine and No. 15 coal from Fenghuangshan coalmine as experimental coal. The latter was reserved at the bottom of the Taiyuan formation in Carboniferous with an average thickness of 2 m. The structure of the coal seam was complex and contained multiple layers of gangue, of which three layers were relatively stable, respectively located in the upper part and the middle part of the coal seam, with the average thickness of 0.01–1.9 m. The pyrite bands and pyrite nodules were visible in the coal seam, part of which existed needle-like or fiber-like pyrite crystals. No.15 coal had spontaneous combustion tendency, which was medium-ash, sulfur-rich, high-heat anthracite. The direct roof of coal seam was limestone, and the direct floor contained bauxite and mudstone. The No. 3 coal seam of the Yuecheng Mine was mainly reserved in the upper part of the Carboniferous Shanxi formation, most of which were semi-bright coals with an average thickness of 6.09 m. The coal seam was stable. No.3 coal had spontaneous combustion tendency which was an anthracite with medium-ash, extra-low sulfur and ultra-high heating value. Minerals appear in the form of stratified coal gangue cracks, and pyrite nodules or fine grain existed in individual fractures. The direct roof of coal seam contained mudstone, silty mudstone, siltstone, and fine, medium sandstones in part. The direct floor contained pink sandstone, mudstone, and fine sandstone in part.

In order to obtain the impact of the difference in coal quality and mineral on PAHs generated in the process of spontaneous combustion, the coal and gangue of the same coal seam were collected respectively for experiments. A total of four samples were collected, 5 kg at each sample collected site, numbered as YCM (raw coal of Yuecheng No. 3 coal seam), YCG (gangue of Yuecheng No. 3), FHM (raw coal of Fenghuangshan No.15 coalseam), FHG (gangue of Fenghuangshan No.15). The raw coal and gangue were directly taken from underground airways. After sampling, they were sealed in plastic bags and taken back to the laboratory, to avoid the samples being oxidized during transportation. Afterwards, 1 kg samples were taken according to the four cone pile method, smashed to 200 meshes in a sealed timing machine. Then the samples were sealed in bags separately. In the end, some broken raw coal and gangue samples were taken for proximate analysis, elemental analysis, calorific value test, and simulation experiments of spontaneous combustion (see Table 1).

3. Experimental

3.1. Experimental equipment

The experimental system includes the coal spontaneous combustion simulation equipment, product pretreatment and analysis and other parts (see Fig. 1). The coal spontaneous combustion simulation equipment is an independent research and development device of China University of Mining and Technology (Beijing). It consists of a reaction tank, a coal sample tank, a furnace body and a thermal insulation layer, a coal tar separator, a product collecting device, an exhaust gas treatment device, a gas pipeline, a thermocouple and so on. It was used to simulate the spontaneous combustion process of coal (gangue). The product pretreatment part consists of Soxhlet extractor, rotary evaporator and chromatography device. The Soxhlet extractor was used to the extract aromatic hydrocarbons after spontaneous combustion gas has been adsorbed by the resin in the adsorbed gas device. Rotary evaporator was used to evaporate the concentrated aromatic hydrocarbon solution. The chromatography device was used for the purification and constant volume removal of impurities, so as to remove polycyclic aromatic hydrocarbons adsorbed in the resin. Shimadzu QP2010 GC-MS was used to determine the type and amount of PAHs.

3.2. Experimental process and parameters

Spontaneous Combustion Experiment. Before the experiment, the pipeline cleaning reagent was acetone. The amount of experimental coal (gangue) was 200 g with the particle size of 4 mesh. The experimental temperature range was from room temperature to 800 °C. Combining the granularity of coal (gangue), the structure of experimental furnace and the critical airflow velocity required for