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Characterization of coal pyrolysis in indirectly heated fixed bed based on field effects

Erfeng Hu [a,b,d], Xi Zeng [b,*], Dachao Ma [c], Fang Wang [b], Yuan Li [d], Erwei Guo [a], Xiaoheng Fu [a]

[a] *School of Chemical & Environmental Engineering, China University of Mining & Technology (Beijing), Beijing 100083, China*
[b] *State Key Laboratory of Multi-Phase Complex Systems, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China*
[c] *School of Environment, Guangxi University, Nanning 530004, China*
[d] *Department of Chemical Engineering, University of New Brunswick, Fredericton E3B 5A3, Canada*

HIGHLIGHTS

- The increased reactor radius raised the coal bed thickness, which decreased the tar yield but raised the light tar faction.
- The char microcosmic study showed the gas flow from pyrolysis sites to central low-temperature coal bed.
- The color variations of quartz verified the change of flow fields, bridging a gap between microcosmic and macroscopic study.

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ABSTRACT

This study is devoted to characterizing the coal pyrolysis performance based on field effects in five fixed-bed reactors with different radiuses. The results showed that the increased reactor radius raised the coal bed thickness, thereby modifying the temperatures and extending the reaction time to reach 500 °C. At a furnace temperature of 900 °C, the increased coal bed thickness from 20 mm to 100 mm decreased the tar yield from 7.24 wt% to 5.62 wt%, while it raised the light tar content from 76.4 wt% to 83.0 wt% in the reactor with internals (reactor B). In contrast, in the reactor without internals (reactor A), the tar yield varied marginally and remained at 4.73 wt% but the light tar content increased from 69.5 wt% to 74.7 wt%. The increased coal bed thickness resulted in an increase in the tar quality but a decrease in the gas HHV (higher heating value) for both reactors. However, with the increase of coal bed thickness, reactor B always provides a higher yield and quality of tar and gas but lower pyrolysis water yield than reactor A, indicating that the internals suppressed the secondary reaction of pyrolysis products and the increase in coal bed thickness did not weaken this advantage of internals. The char HHV located in the center of the reactor with internals was higher than that of the reactor without internals; this was postulated that the pyrolysis products escaped from the central low-temperature coal bed, which enhanced carbon deposition. As expected, EDS results proved the postulation and showed that the char in the center of the reactor B had more carbon species. In addition, the color changes of quartz sand in the before and after tests first verified the flow field of pyrolysis products in phenomenology.

1. Introduction

Due to the rapid consumption of energy and the constant depletion of fossil fuels, the exploration of a supplementary fuel oil becomes progressively urgent. Now, much research has been devoted to the development of coal pyrolysis, and are aimed at developing a high-efficiency clean technology for converting coal volatiles into oil [1–4].

In the past centuries, many technologies have been developed for coal pyrolysis, mainly direct-heating pyrolysis technology and indirect-heating pyrolysis technology. The former, such as Toscoal technology [5], DG technology [6,7], LFC technology [8,9], COED technology [10,11], and Encoal technology [12], is mainly heated by solid or gaseous heat carriers, which is characterized by high tar yield because of rapid heating rate. However, the limitation is poor oil quality caused by the entrainment of fine particles and higher content of heavy components. On the other hand, although partial indirect heating technologies, such as W-D technology, Koppers technology [13] and MRF technology [14,15] can

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* Corresponding author.
E-mail address: xzeng@ipe.ac.cn (X. Zeng).

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effectively resolve the limitations mentioned above, they still need to overcome the problems of the slow heating and low heat transfer efficiency. Based on this concept, the fixed-bed reactor with newly designed internals has been proposed by Zhang et al., and this newly configured reactor raised the tar yield and quality by changing the flow field of pyrolysis products [16,17]. Siramard et al. [18]. reported that the reduced pyrolysis pressure in this reactor reduced heat transfer, but raised shale oil yield. This reactor was also adopted on oil shale pyrolysis by Lai et al. [19]and Lin et al. [20], illustrating a similar improvement on yield and quality of oil shale. However, the effect of bed thickness on pyrolysis behavior in this reactor has not been studied. In addition, the preceding studies all proposed a theoretical conjecture of flow field of pyrolysis products, whereas the flow filed has never verified in phenomenology.

The extensively reported works were focused on the study of coal rank [21], coal particle diameter [22], coal mineral composition and content [23], different atmosphere [24] heating rate [25], coal composition [26,27], and operation conditions [27–31]. In the only reported work of coal bed thickness effects, Solomon simulated that increasing the bed thickness raised tar-char secondary reactions which diminished tar yield [30]. Mazumdar et al. found that the increased thickness exacerbated the degree of interaction and inhibition of tar formation, which further resulted in a higher coke yield and lower tar yield in industrial high-temperature carbonization [31]. Based on literature review, the coal bed thickness was an important factor to affect the carbonizations. Thus, the coal bed thickness effects on pyrolysis behavior and macroscopic verification of flow field were further investigated.

This work aims to study the coal bed thickness effects on pyrolysis behavior in newly configured fixed-bed reactors with internals, as well as the characteristics of tar components and char. Those results were also compared with the results from the reactors without internals. In addition, the flow field effects were first verified by analyzing the color changes of quartz sands in the before and after tests through the quartz layer.

2. Experimental

2.1. Materials

The sample adopted was a kind of sub-bituminous coal from Yilan, Heilongjiang province of China. Prior to each experiment, coal sample was crushed below 5 mm and then stored in a sealed bag. Table 1 lists the main characteristics of tested coal sample, showing a relatively high ash content (42.08 wt%).

2.2. Apparatus and methods

Fig. 1 shows the schematic diagram of two kinds of reactors for the pyrolysis tests (reactor A and B). Reactor A did not have any internals, while four metallic plates and a central gas collection pipe (internals) were mounted in reactor B. The plates in reactor B were contacted with the heated reactor wall and at 90° to one another. Both reactors were made of 304-type stainless steel. The reactors with the same height but different inner radiuses (20 mm, 40 mm, 60 mm, 80 mm and 100 mm) were used in this study. Prior to each experiment, the furnace (1) temperature was heated to 900 °C firstly, and then the reactor connected to the gas collection system (5, 6, 7, 8) was quickly placed into the furnace. The produced pyrolysis gas was measured by a wet gas meter (11) for total volume, and then analyzed by gas chromatography after passing through the gas purification system (13,14). To keep the highest tar yield at the shortest reaction time, all the experiments were terminated when the temperature of central coal bed reached 500 °C. After each experiment, the whole condenser pipe was washed by warm acetone and then mixed with cool acetone in bottles (6–8); the acetone was then evaporated and removed to get the tar. The water in the bottle (5) was separated by oil-water delamination and the moisture content in the tar was determined by the water-toluene distillation method. The gas and char were also collected to analyze their yields.

The study also verified the flow field of pyrolysis gas through the quartz layer in both 40-mm-radius reactors. Specifically, a cylindrical steel net with the radius of 30 mm and mesh of 40 was put into reactors firstly, and then the coal and quartz sand were loaded into reactors, respectively. A1, A2, B1 and B2 represented four types of the experiments, and its filling patterns of quartz and coal are shown in Fig. 2, respectively. For A1 and B1, the coal was filled at the center of reactor while the quartz sand was loaded at the outer side of the reactor; For A2 and B2, they showed an opposite filling patterns. The experiments were conducted with the procedures mentioned above.

2.3. Products analysis

The sampled gas was analyzed by a micro GC (Agilent 3000A) to detect the concentrations of gas species including H_2, CH_4, CO, CO_2, C_2H_4, C_2H_6, C_3H_6 and C_3H_8. The dehydrated tar was analyzed in a simulated distillation GC (Agilent 7890) to determine the distribution of light tar fraction. The components below the boiling points of 360 °C were defined as light tar. The calorific value of char was measured using an oxygen bomb calorimeter (Shanghai Jichang XRY-1B), and the char surface morphology was examined by an atomic force microscope (AFM) equipped with a scanning electron microscope (SEM JSM-6700 F). The yields of pyrolysis products (wt.%) were calculated based on the dry weight of coal; the relative errors of the experiments were less than 3%.

3. Results and discussion

3.1. Heating characteristics for coal

Fig. 3 shows the heating curves of coal in the center of reactor A and B under different coal bed thickness conditions. The increased coal bed thickness extended the reaction time for the central coal to reach 500 °C for both reactors; the reaction time in reactor B (with internals) was less than that of reactor A under the same coal bed thickness. When the coal bed thickness was 20 mm, the coal reaction times for reactor A and B were 19.5 min and 25 min respectively; the reaction times prolonged to 143.5 min and

Table 1
Proximate and ultimate analyses for the tested Yilan coal

Proximate analysis (ar, wt%)				Ultimate analysis (daf, wt%,)					G-K (d, wt%)[†]	HHV(kJ·kg-1)
Mt	A	V	FC	C	H	N	S	O[*]	Tar	Coal
4.61	42.08	27.24	26.07	69.15	7.28	2.00	0.80	20.77	7.88	16214

[*] Determined by element mass balance (as received);
[†] Tar yield from Gray-King assay test.