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Coal pyrolysis and its mechanism in indirectly heated fixed-bed with metallic heating plate enhancement

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HIGHLIGHTS

- The tar yield and light fraction increased then decreased when increasing the number of plates.
- Working mechanism of metallic plates was proposed and verified by an in-house design.
- An excessive number of plates resulted in aggravating the cracking of primary products.

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ABSTRACT

A metallic plate has been devised to enhance the heat transfer; this in turn increases the yield and quality of coal tar (Zhang et al., 2013). This work is devoted to investigating the pyrolysis of Yilan subbituminous coal and developing a working mechanism for an indirectly heated fixed bed reactor with different number of metallic plates. Increasing the number of metallic heating plates enhanced the heat transfer from the high-temperature reactor wall to the low-temperature central coal layer and thereby shortened the reaction time. Meanwhile, the yields of tar and light fraction were increased then decreased. The results of the pressure drop experiment demonstrated that the metallic plates weakened dense stacks of coal particles and raised the particle interstices, and therefore lowered the gas diffusion resistance. It verified that many gaseous pyrolysis products escaped from the central low-temperature coal layer, indicating the suppressed secondary reactions to the primary products. When the number of plates was 8, the correspondingly added high-temperature surface was 113%; it achieved the optimal matching between the secondary reactions of pyrolysis products and the fields of the temperature as well as the gas flow inside the reactor, and it also had the highest tar yield (6.60 wt.% dry basis) and the maximum content of light tar (below the boiling points of 360 °C) of 76.4 wt.% and C4–C9 components of 43.52 wt.%. The BET results showed that the char had the biggest surface area and volume for this case with 8 plates. Consequently, adding suitable metallic plates into the indirectly heated fixed bed reactor obviously enhances the heat transfer and also achieves higher yield and quality of tar.

1. Introduction

Due to large quantities of low-rank coal resources but limited reserves of oil and gas, advanced coal utilization technologies have attracted more and more attention from researchers. Pyrolysis is the critical core technology for cascade utilization of low-rank coal; it is becoming a promising research focus for energy systems, especially in China.

A large number of fundamental and technical studies of coal pyrolysis have been done for decades. Many fundamental aspects, including the coal rank [2], coal particle diameter [3,4], coal mineral composition and content [5], pyrolysis atmosphere [6], as well as the heating rate [7,8], coal composition [9,10], and operating conditions [11–14], were extensively investigated. In the technical aspects, mostly the existing coal pyrolysis technologies based on solid heat transfer (char or hot ash), such as Toscoal [15]and DG [16,17], have been examined. There are also ones using gaseous heat carrier, such as COED [18,19], Encoal [20] and LFC [21,22]. There are drawbacks, however, that include complex process,

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unstable operation and especially poor oil quality in those technologies, which limit their expansion. Because of no heating medium, relatively simple process and good oil quality, indirect heating technology may be a good way to upgrade low rank coal if its low heat transfer efficiency can be solved.

Elevating the heating temperature, accelerating the heating speed or reducing the material particle size accelerates the heat transfer. Khan [23] reported that in an indirectly heated fixed-bed reactor, the tar yield increased when the heating temperature rose from 500 °C to 649 °C, whereas the tar yield decreased with increased heating temperature above 650 °C [1]. Peters and Bertling [24] found the tar yield increased when the heating rate was raised. Hu et al. [25] found fine particles facilitated heat transfer but aggravated secondary reactions, resulting from the low gas permeability. Zhang et al. [1] found the metallic plate accelerated the heat transfer and raised the tar yield in fixed-bed reactors. Lin et al. [26] determined that appropriately increasing the number of metallic plates reinforced the heating transfer of oil shale and also increased yield and quality of shale oil in a newly configured reactor with internals. Nonetheless, little work has been done to evaluate how the number of metallic heating plates in coal pyrolysis would affect the pyrolysis products. What is more, the investigation of the working mechanism is absolutely necessary for the coal pyrolysis process.

This study aims to investigate the coal pyrolysis behaviors in an indirectly heated fixed-bed reactor with different number of metallic plates, and the components of tar and micro-characteristics of char are analyzed. In addition, the working mechanism of plates is also studied and verified by an in-house design.

2. Experimental

2.1. Materials

The sub-bituminous coal sample from Yilan, Heilongjiang, China, was crushed into sizes below 5 mm for tests and stored in a sealed bag for later use. The main property data for the coal sample are listed in Table 1. The coal had high ash content, and its volatile content was about 27 wt.%. The Gray-King gave a tar yield of 7.88 wt.% against dry coal mass.

2.2. Apparatus and procedure

The left inset of Fig. 1 exhibits the fixed-bed reactor and metallic plates used in this paper. They were both made of 304-type stainless steel. The right inset of Fig. 1 presents a schematic diagram of the test system. It consisted mainly of an electric furnace (1), a reactor (2), a pressure gauge (3), a cooling and adsorption system (4–8), a filter (9), a vacuum pump (11), a sodium bicarbonate washing bottle for removing sulphureted hydrogen (13), and a silica gel bottle (14). Cotton wool was used to insulate both reactor and furnace. The reactor was 45 mm in inner diameter and its total heating surface area which was provided by reactor heating wall was 254 cm^2 for coal samples. The metallic plates were perpendicular to the reactor wall and mutually at the same angle. The plate was 15 mm in width and 120 mm in height. As shown in Table 2, corresponding to the number of 0, 4, 8 and 12 of the added metallic plates, the added high-temperature heating surface reached 0, 57%, 113% and 170% of the reactor heating wall, and those were denoted as a, b, c and d respectively.

The results of the previous study have demonstrated that Yilan coal had the highest yield of tar and gas when the central temperature reached 500 °C [1]. Consequently, each test was ended when the same central position reached 500 °C. As illustrated in Fig. 1, coal was fed into the reactor and then the reactor was connected with gas collection and purification at the beginning of the experiment; at the end of experiments, cooled char was weighed to calculate the yield. The product in the collection bottle (5) was also weighed, and then the water in the bottle (5) was poured into a clean beaker. The pyrolysis products attached to the condenser (4) were washed by acetone, and then mixed with acetone solution from the bottles (6, 7, 8). The acetone in the solution from the bottles (6, 7, 8) was removed by a vacuum rotary evaporator. The recovery tar was mixed with the preceding tar in the collection bottle (5). The moisture content in tar mixture was further determined and water-free tar yield was obtained by using toluene as azeotrope according to the standard method. The moisture contained in tar mixture and the preceding water poured into a clean beaker were calculated as the pyrolysis water. The gas volume was recorded on a wet gas meter to calculate pyrolysis gas yield. The experiments were repeated and the relative errors were less than 3%. Experimental procedures were substantially similar to those of Zhang [1].

This study also measured the pressure drop across a char or a coal bed in a N_2 flow of different fluxes. A quartz tube with a sintered orifice was used as gas distributor and was 40 mm in diameter and 400 mm in height. A total of 130 mm in height of coal or char was loaded into the tube to measure the pressure drop across the material bed at different N_2 fluxes (see Fig. 2). Then the metallic plates were loaded in the tube with coal or char and measurement of the pressure drop was conducted by varying the number of plates.

2.3. Products analysis

The micro GC (Agilent 3000 A) detected the composition of sampled non-condensable gases. Tar in experiments was removed in water and collected into a vial. Then, the dehydrated tar entered the micro GC (Agilent 7890), which can determine fraction distribution for diverse boiling points. The light tar consisted of those components with boiling points below 360 °C. The composition of dehydrated tar was also measured by using a GC–MS spectrometer (Shimadzu QP 2010 Ultra). The injector and detector reached a temperature of 280 °C, and the column of GC was heated to 50 °C in 5 min and further to 280 °C at 6 °C/min. Finally the column was kept at 280 °C for 10 min. The scanning range was from 20 to 900 m/z and the delay time of solvent was 1.7 min. The relative content of components was evaluated with the peak area percentage, i.e., the peak area proportion to total peak area.

The calorific value of char was measured with a Shanghai Jichang XRY-1B oxygen bomb calorimeter. The char surface morphology was examined by an atomic force microscope (AFM) equipped with a scanning electron microscope (SEM JSM-6700 F,

Table 1
Proximate and ultimate analyses for the tested Yilan coal.

Proximate analysis (ar, wt.%)				Ultimate analysis (daf, wt.%)					G-K (d, wt.%)[b]	HHV($kJ\cdot kg^{-1}$)
Mt	A	V	FC	C	H	N	S	O[a]	Tar	Coal
4.61	42.08	27.24	26.07	69.15	7.28	2.00	0.80	20.77	7.88	16,214

[a] Determined by element mass balance.
[b] Tar yield from Gray-King assay test.