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Journal of Analytical and Applied Pyrolysis



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

Coal pyrolysis in a fluidized bed reactor simulating the process conditions of coal topping in CFB boiler

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ARTICLE INFO

Article history: Received 14 September 2010 Accepted 18 February 2011 Available online 26 February 2011

Keywords: Coal topping CFB boiler Fluidized bed Reaction atmosphere Pyrolysis oil TG-FTIR

ABSTRACT

Simulating the conditions of pyrolytic topping in a fluidized bed reactor integrated into a CFB boiler, the study was devoted to the reaction fundamentals of coal pyrolysis in terms of the production characteristics of pyrolysis oil in fluidized bed reactors, including pyrolysis oil yield, required reaction time and the chemical species presented in the pyrolysis oil. The results demonstrated that the maximal pyrolysis oil yield occurred on conditions of 873 K, with a reaction time of 3 min and in a reaction atmosphere gas simulating the composition of pyrolysis gas. Adding H₂ and CO₂ into the reaction atmosphere decreased the pyrolysis oil yield, while the oil yield increased with increasing the CO and CH₄ contents in the atmosphere. TG-FTIR analysis was conducted to reveal the effects of reaction atmosphere on the chemical species present in the pyrolysis oil. The results clarified that the pyrolysis oil yield reached its maximum when the simulated pyrolysis gas was the reaction atmosphere, but there were slightly fewer volatile matters in the pyrolysis oil than the oil generated in the N₂ atmosphere. All of these results are expected not only to reveal the composition characteristics of the pyrolysis oil from different conditions of the coal topping process but also to optimize the pyrolysis conditions in terms of maximizing the light pyrolysis oil yield and quality.

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1. Introduction

"Coal topping" (pyrolytic topping) process was proposed by Yao and Kwauk [1,2] to achieve high-value utilization of coal consumed in CFB boilers. As shown schematically in Fig. 1, light liquid product in this process is produced by flash pyrolysis of coal in a pyrolyzer integrated into a circulating fluidized bed boiler which burns the pyrolysis-generated char to generate heat and electricity. Previous studies showed that the pyrolysis is necessary to proceed with rapid heating and in turn the quick separation (from solids) and quench of the gaseous product in order to minimize the secondary reactions like cracking and polymerization [2,3].

A few, although limited studies have been done regarding pyrolytic topping. Wang et al. [4] found that downer is a suitable reactor for implementing the pyrolysis of coal through mixing with the hot ash particles during their fall in the reactor via gravity. Those authors further investigated the influences of pyrolysis temperature and particle size on the topping performance [5,6]. Because the coal particle residence time is short (only a few seconds), the downer reactor only adapts to coal in micrometers. Moving bed is another kind of reactor used to implement the pyrolytic topping, and Bi [7] employed this reactor to integrate the coal pyrolysis with a riser combustor. Comparing to the downer reactor, a special effort is needed to scale up the reactor to achieve high-efficiency mixing with coal particles for industrial application. Fluidized bed allows easier scale-up and also adapts wide-size particles (<10 mm), and it should be highly appropriate for pyrolytic topping. However, almost no related study has been conducted before under this concern. Thus, this work is devoted to investigating the coal pyrolysis characteristics in terms of pyrolysis oil yield and quality under conditions simulating the pyrolytic topping process. The effects of temperature, bed height, reaction time and reaction atmosphere were studied to deepen the understanding of coal pyrolysis and to provide fundamental data for the process and operation optimizations of the pyrolytic topping.

2. Experimental

2.1. Apparatus and test procedure

The experiments were performed in a quartz-made fluidized bed reactor of 0.06 m in diameter and 0.7 m in height. The schematic diagram of the experimental apparatus is shown in Fig. 2. The coal particles were fed from the top of the bed into the reactor in which quartz sand particles of 212–380 μ m were fluidized. The height of

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^{0165-2370/\$ -} see front matter © 2011 Elsevier B.V. All rights reserved. doi:10.1016/j.jaap.2011.02.013

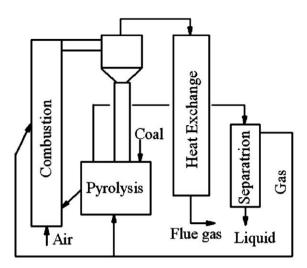


Fig. 1. Principle of the coal pyrolytic topping process in CFB boiler.

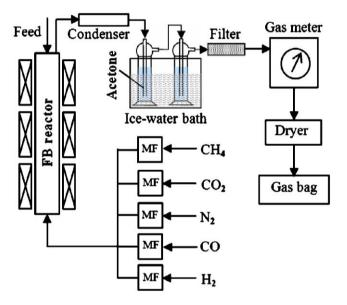


Fig. 2. Schematic diagram of the adopted experimental apparatus.

the sand bed in the reactor was adjusted according to the experimental needs. A three-zone electric furnace heated the reactor and the heating conditions for each zone could be independently controlled. A thermocouple was immersed in the quartz sand bed to monitor and measure the temperature of the fluidized bed. The reaction atmosphere (N₂ or mixture of H₂, CO, CO₂ and CH₄) was formed by mixing gases from different cylinders and the flow rate was kept at 1.15 m/min (2 times of minimum fluidization velocity) to achieve the full fluidization of the quartz sand particles. The particle sizes of the tested lignite were 4–6 mm, and Table 1 shows the major properties of the tested coal.

Table 1

Proximate and ultimate analyses of the tested coal.

Proximate analysis [db-wt.%]	Ultimate analysis [db-wt.%]
Water (arrival base): 1.7	C: 65.6
Volatile: 31.2	H: 4.1
Ash: 18.2	S: 0.6
Fixed C: 48.9	N: 1.1
LHV [MJ kg ⁻¹] 24.96	O: 9.7

Table 2

Absorbance [cm ⁻¹]	Function group	Characteristic chemicals
1355–1395, 1430–1470	-CH ₃	Methyl
1405-1465	-CH ₂	Methylene
1500	C=C	Single ring aromahydrocarbon
1740	C=0	Acid ketone
2800-3100	CH ₄	Methane
2000-2250	CO	Carbon monoxide
2250-2400	CO ₂	Carbon dioxide
2920	-CH ₃ , =CH ₂ , ≡C-H	Aliphatic
3500	-O-H(a)	Phenolic hydroxyl
3650	-O-H(b)	Alcoholic hydroxyl

The reactor was first heated to 673 K before the fluidizing gas (N₂) was introduced into the reactor. The bed was heated to the desired temperature in N₂ atmosphere and then the fluidizing gas was switched to the required reaction atmosphere. When the bed temperature in the new atmosphere reached the specified steady value, 10 g of lignite were added into the reactor from the bed top through a valve-hopper. The generated pyrolysis gas was cooled immediately in a water cooler and then washed with water and acetone in succession cooled via an ice-water bath. The volume of the gas was measured by a wet gas meter after the washing bath. After passing through a filter and drier further, the cleaned gas was sampled at the end of the gas line at an interval of about 20 s. At the end of gas sampling, the gas from the reactor was switched to a bypass line to vent without passing through the above-mentioned gas cooling and washing vessels. Therefore, the time to sample the gas also represented the reaction time measured for the pyrolysis, which was usually a few minutes after the coal feeding. The sampled gas was analysed using a micro GC (Agilent 3000) to determine its composition.

The liquid collected from washing the cooler and pyrolysis gas was treated to recover pyrolysis oil (tar) through filtration and remove both acetone and water. The collected liquid was first treated in an atmospheric rotary evaporator at 318 K and then in a vacuum oven at 318 K. The quality of the pyrolysis oil was determined by a thermal gravity analyzer integrated with a Fourier transform infrared spectrometer (FTIR).

2.2. Analysis approach and mass balance

A micro GC was used to measure the molar concentrations of H_2 , O_2 , N_2 , CO, CO_2 and hydrocarbons up to C3 in the gaseous product. In order to gain composition information of pyrolysis oil, a thermo gravimetric analyzer (Netzsch STA 449C) coupled with a FTIR (i.e., TG-FTIR) was employed to analyse the recovered pyrolysis oil. In the TG-FTIR analysis, the pyrolysis oil sample was heated from 303 K to 1173 K at 30 K/min in TG. Nitrogen at 80 mL/min was adopted to carry the evolved volatile gas from TG to the gas cell of FTIR heated to 573 K. The FTIR analysis was conducted at TG temperatures between 373 K and 1273 K. The resulting intensity of FTIR spectrum was mass-normalized to eliminate the influence of sample mass. Table 2 lists the characteristic peaks in FTIR spectra and their implicated functional groups. The yield of pyrolysis oil (Y_{oil} , wt.%) and the production rate of pyrolysis gas (Y_{gas} , L/g) with a dry ash-free basis were calculated with

$$Y_{\rm oil} = \frac{m_{\rm oil}}{m_{\rm coal} \times (100 - M_{\rm ad} - A_{\rm ad})} \times 100\%$$
(1)

and

$$Y_{\rm gas} = \frac{22.4 \times \sum_{i} \int_{0}^{t} F_{\rm mt} C_{i} dt}{m_{\rm coal} \times (100 - M_{\rm ad} - A_{\rm ad})} \times 100\%$$
(2)

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