Fuel Processing Technology 116 (2013) 35–43

Contents lists available at SciVerse ScienceDirect

Fuel Processing Technology

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

Lignite upgrading by multi-stage fluidized bed pyrolysis

Qi Zhou [a,b], Tao Zou [a,c], Mei Zhong [d], Yuming Zhang [a,b], Rongcheng Wu [a], Shiqiu Gao [a,*], Guangwen Xu [a,*]

[a] State Key Laboratory of Multiphase Complex Systems, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China
[c] China University of Mining and Technology, Beijing 100083, China
[d] Department of Chemistry and Chemical Engineering, Xinjiang University, Xinjiang 830046, China

ARTICLE INFO

Article history:
Received 4 February 2013
Received in revised form 17 April 2013
Accepted 18 April 2013
Available online xxxx

Keywords:
Multi-stage fluidized bed
Lignite upgrading
Pyrolysis
Gasification
Lignite

ABSTRACT

This study is devoted to demonstrating experimentally the technical advantages of the multi-stage fluidized bed pyrolysis for upgrading lignite. A Chinese lignite was pyrolyzed and partially gasified in a three-stage laboratory-scale fluidized bed, with an overflow standpipe between its neighboring stages, to clarify the improvement on the pyrolysis product quality by increasing the number of the stages. While the bottom stage had the highest temperature of about 900 °C for fuel gasification, the upper stage had temperatures of 550–650 °C for coal pyrolysis. The multi-stage fluidized bed was operated with a continuous feed in the modes with one to three stages. The resulting yields of gas and tar were higher, whereas the yield of char was lower for the operations with multiple stages. The produced CO, H_2 and CH_4 in the two- and three-stage modes were more than that in the single-stage mode, having thus the higher gas heating value as well. The tar from the three-stage fluidized bed pyrolysis contained more light oil, and it plus phenol oil reached 99.5 wt.% of the tar. The char produced in the multi-stage pyrolysis showed the higher thermal stability in terms of its higher ignition temperature and suppressed spontaneous combustion propensity.

1. Introduction

Lignite has moisture contents of 30–50 wt.% and is easy to burn spontaneously during processing and storage. Upgrading lignite aims at not only decreasing its moisture content but also suppresses its propensity of spontaneous combustion. Thus, coal pyrolysis is the most common way to convert lignite into the high-rank fuel like the other types of coal.

By far, many pyrolysis technologies have been developed to upgrade lignite and meanwhile coproduce tar in some of the technologies. The well-documented typical technologies have the Toscoal [1,2], LFC [3], COED [4,5], Lurgi [6], Garrett [7], ECOPRO [8] and DG [9]. Some of these have been commercially demonstrated at capacities of above 1000 t/d coal, but now there is not any pyrolysis plant in commercial running. Reported technical problems are all related with the simultaneously generated tar. The typical problem is the high fraction of heavy components such as pitch in the produced tar, which means not only the low quality of tar and its difficulty in downstream utilization but implies as well the troubles in running continuously the pyrolysis process due to the deposition of heavy tar on the ducts after the pyrolyzer. Nonetheless, the Lurgi, COED and ECOPRO processes have been reported to produce tar with more light fractions.

The Lurgi moving bed gasifier, which incorporates pyrolysis in fact, produces tar with high light fraction through pyrolyzing the fed coal with temperature rising gradually in the circumstance of gasification gas coming from its lower char gasification section. But this gasifier can adapt only to lump coal in sizes above 6 mm and its tar yield is relatively low [10]. The COED process is adaptive to powder coal below 3 mm and it pyrolyzes coal in four successive fluidized bed (FB) reactors with ascending temperatures that are particularly designed for implementing coal drying, mild pyrolysis, deep pyrolysis and finally gasification. Like in Lurgi gasifier, the produced gas and solid coal/char flow countercurrently so that the produced tar can have high light fractions of oil (boiling points below 360 °C) [11,12]. The ECOPRO process uses an entrained bed reactor to treat pulverized coal in sizes below 50 μm and it consists of an upper section for fast pyrolysis and a bottom section of ash-melting char/coal gasification so that the coal pyrolysis occurs also in the gasification-generated gas. With higher processing temperatures at H_2 rich syngas atmosphere, the generated tar was reported to contain about 90% light fractions [8].

Literature studies found that the upgraded lignite had higher propensity of spontaneous combustion when there were enough volatile matters remaining in the upgraded coal [13,14]. Olayinka et al. [15] reported that the thermally treated lignite at higher temperatures hardly occurred the spontaneous combustion. Inorganic species and additives may promote spontaneous combustion of low rank coal [16]. Faúndez et al. [17] found that the ignition temperature was inversely correlated with the reactivity of the upgraded coal. Liu et al. [18] have investigated the gasification reactivity of three types of chars in CO_2 atmosphere in a fluidized bed, finding that longer pyrolysis

* Corresponding authors. Tel.: +86 10 82544885; fax: +86 10 82629912.
E-mail addresses: sqgao@home.ipe.ac.cn (S. Gao), gwxu@home.ipe.ac.cn (G. Xu).

0378-3820/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.fuproc.2013.04.022

time led to lower gasification reactivity of the produced char. Zanzi et al. [19] found that the char from fast pyrolysis had higher gasification reactivity than that produced by slow pyrolysis. Low reactivity due to high pyrolysis temperature (1475 K) was reported also by Radovic et al. [20]. Krister et al. [21] revealed that the secondary interaction of tar with char reduced the reactivity of char gasification with steam. All of these show that the thermal stability of the produced char should be improved by means of lowering the volatile matters and inorganic components in the char, prolonging the pyrolysis time, increasing the pyrolysis temperature and promoting the interaction between char and tar.

We proposed the use of a multi-stage fluidized bed, with ascending temperatures from the top stage to the bottom stage, to produce high-quality tar and high thermally stable char through coupling the coal pyrolysis and partial char gasification in a single reactor. This article is devoted to demonstrating the technical advantages of the multi-stage fluidized bed over the usual single-stage fluidized bed for lignite upgrading by pyrolysis. In principle, the multi-stage fluidized bed can have the similar temperature gradient as the Lurgi gasifier from its top to bottom, but the multi-stage fluidized bed can treat coal with sizes below 6 mm. In this article, a three-stage laboratory-scale fluidized bed was used to test the upgrading of lignite by pyrolysis and partial gasification and to demonstrate its technical superiority.

2. Experimental

2.1. Apparatus and fuel

Fig. 1 presents a schematic diagram of the employed experimental setup that is consisted mainly of a screw feeder, a three-stage fluidized bed reactor, an electric furnace, a gas supplying system, a gas–solid separator, a char collector and a tar recovery system. All the operating parameters including gas flow rate, temperature and pressure were monitored and sampled by a data acquisition system. The reactor was made of a SUS310S stainless steel tube of 102 mm in inner diameter. Hereafter, the bed stages from the bottom to top is named the first, second and third stages, respectively. The height of the reactor was 1800 mm above the distributor of the first stage. While the third stage was 1000 mm high, the other two stages were 400 mm high for each. The used distributor for the second and third stages was a perforated plate with an overflow standpipe, which had an opening area ratio of 3.3% and the orifices of 2 mm in diameter. The standpipe worked to transfer the coal or char from an upper stage to its lower stage, and its diameter was 20 mm. By adjusting the height of the standpipe above the distributor, the height of particles in the second and third stages was kept at 120 mm. Dried coal was fed into the third stage from a point about 650 mm above the third distributor. A perforated plate without an overflow standpipe was used as the distributor for the first (bottom) stage, which had a 1.0% opening area ratio and the orifices of 1.2 mm in diameter. An inclined overflow pipe was mounted outside the reactor at 120 mm above the first distributor to discharge char during continuous operation.

The multi-stage fluidized bed can be operated in the modes with one to three stages to investigate the influence of the number of stages, as is shown in Fig. 2. That is, the bottom stage was used for the single-stage operation, while the first and second stages were adopted to form the two-stage operation mode. Beneath the bottom (first) stage is a gas preheater filled with inert Al_2O_3 ball of 3–4 mm in size, which can preheat the fluidizing gas to about 600 °C. Three electric furnaces were used to heat the three stages and adjust independently their temperatures that were monitored with K-type thermocouples.

The tested lignite was from Inner Mongolia Autonomous Region of China, and it was crushed and sieved to sizes of 0.2–0.5 mm. Before

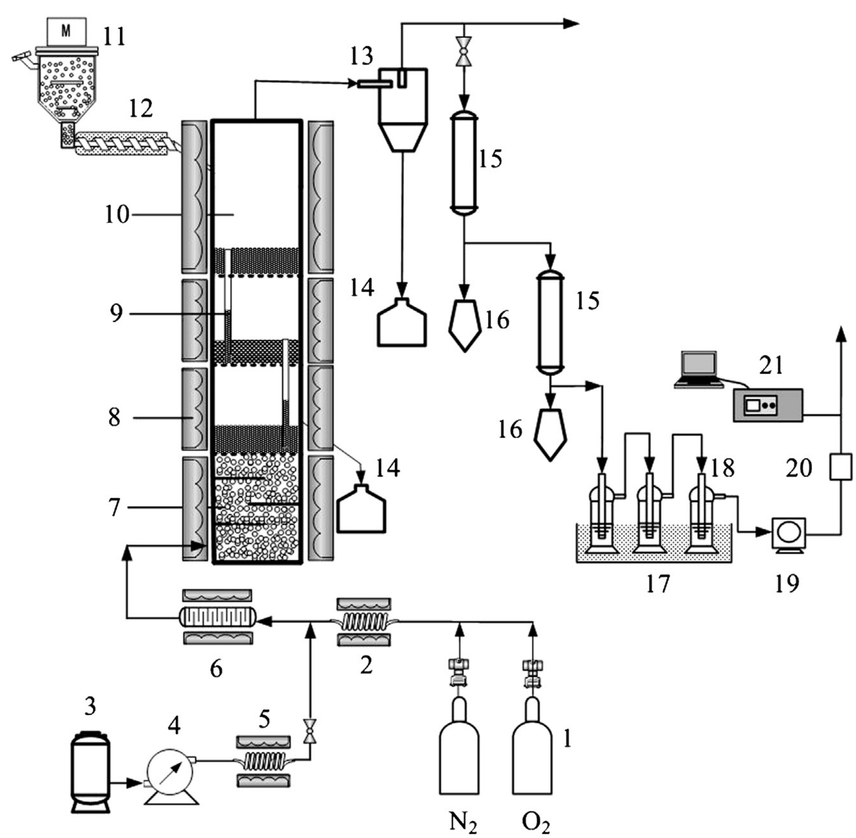

Fig. 1. A schematic diagram of experimental system. (1) Gas cylinders; (2) Gas preheater; (3) Water tank; (4) Plunger pump; (5) Steam generator; (6) Gas mixer and preheater; (7) Preheater; (8) Electric furnace; (9) Overflow standpipe; (10) Reactor; (11) Coal hopper; (12) Screw feeder; (13) Cyclone; (14) Char receiver; (15) Condenser; (16) Tar collector; (17) Ice-water bath; (18) Acetone trap; (19) Wet gas meter; (20) Gas bag; and (21) Micro GC.