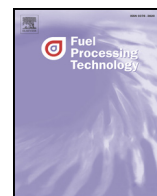




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Characteristics and application of co-pyrolysis of coal/biomass blends with solid heat carrier

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ABSTRACT

A series of experiments on co-pyrolysis of subbituminous coal/corn stalk blends with solid heat carrier (boiler ash) were carried out in a bench-scale moving bed pyrolyzer with different biomass blending ratios at different temperatures. Deviation of gaseous, liquid and char products yields between experimental values and calculated ones was discussed. Some characteristics of char and char-ash after co-pyrolysis were investigated, such as elemental analysis, heat value, ash melting point and ash composition. Results indicated that with increasing blending ratio of corn stalk and temperature, both gas and tar yields increased. Gas heat value decreased with increasing corn stalk blending ratio while increased with rising temperature. Compared with coal pyrolysis alone, more light-oil and water were found during the co-pyrolysis process. The char-ash showed little impact on boiler combustion ash at the blending ratio of biomass less than 30%. Application of co-pyrolysis in a poly-generation system was evaluated environmentally and economically.

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1. Introduction

Biomass is getting increasing attention as a potential resource of renewable energy due to high yield of volatile matter, possibility of being converted into liquid fuels and chemicals, low contents of sulfur and nitrogen as well as zero net emission of CO₂. However, the drawbacks of biomass, such as low energy density, seasonal, difficult to collect and transport etc., limit its utilization greatly. One of the most promising solutions is to co-utilize with coal, such as co-combustion [1–4], co-gasification [5–8] and co-pyrolysis [8,9]. These solutions cannot only avoid the disadvantages of biomass and coal, but also exert the advantages of them simultaneously. Since co-pyrolysis process of coal and biomass can produce gaseous and liquid products with high added value, its research has been paid more and more attention in recent years. Many researchers used different fuels (such as sawdust [10,11], legume straw [12], lignite [13–17], and bituminous coal [18]), and different reactors (such as TGA [19–22], fluidized bed reactor [8], fixed-bed reactor [11], and free fall reactor [12]) under various operating parameters (such as temperature [23], heating rate [24], blending ratio, particle size, and contacting way of particles) to study the co-pyrolysis behaviors focusing on product distributions and product characteristics, as well as the possible existed synergetic effects. Although the conclusions are conflicting, but there are some certain laws: under the conditions of fast heating rate and good contact of

coal/biomass particles, the synergistic effects are obvious; in contrast under the condition of slow heating rate and fluidized bed reactor, the synergistic effects are rarely found. But so far, there is scarcely report on application of co-pyrolysis of coal and biomass, especially by using coal-ash from a boiler as solid heat carrier.

In order to realize the efficient and clean utilization of coal, a poly-generation process, which was coupled circulating fluidized bed (CFB) combustion (15 t/h coal and gangue feeding, 75 t/h steam output) with coal pyrolysis (5 t/h coal feeding), was finished in a pilot-scale [25]. In the poly-generation process, a pyrolyzer was set beside the CFB boiler and connected with it. High-temperature ash from the CFB boiler was quantitatively transported into the pyrolyzer to provide heat for coal pyrolysis, getting gas and tar. The char produced in the pyrolyzer was returned to the CFB boiler for combustion, getting heat or power. Thus, the poly-generation of gas, tar, heat and power can be obtained simultaneously in the process. The coal pyrolysis with the ash has been researched previously in our laboratory [26–28]. Because of the high content of volatile matter in the biomass, if adding some biomass into the pyrolyzer to co-pyrolyze with coal, it may produce more valuable gas and tar, which can improve the utilization of biomass and the economic benefits.

Co-pyrolysis behaviors of coal and biomass with the high-temperature boiler ash as solid heat carrier were investigated in this work. The yields of gas, liquid, char and their characteristics were studied at different biomass blending ratios and at different pyrolysis temperatures since they were the most important parameters in the fast co-pyrolysis process. In addition, the application of co-pyrolysis in

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a poly-generation system was evaluated environmentally and economically. The results may provide useful information for the application of coal/biomass co-pyrolysis system.

2. Experimental

2.1. Materials

The coal used was Fugu subbituminous coal (FSBC) from Fugu coal field, one of the main areas producing subbituminous coals in China. The biomass used was corn stalk (CS) from Taiyuan Shanxi because corn was one of the main food crops in the two provinces (Shaanxi and Shanxi) and a great deal of corn stalks were produced annually. The air-dried coal samples were crushed to the size of below 4.0 mm. In order to feed conveniently the air-dried corn stalks were first crushed to powder, and then granulated to cylinder form with diameter of 2.0 mm and length of 6.0 mm. The proximate analyses were performed according to GB/T 212–2001 (Chinese standards). The ultimate analysis was carried out using a CHNS/O Vario Micro Cube elemental analyzer. The results of the proximate and ultimate analyses of coal and granulous corn stalk particles are listed in Table 1. To eliminate the effect of moisture contained in the raw materials, prior to each test sufficient amounts of fuel samples were dried at 105 °C for more than two hours and then stored in a desiccator to prevent extra absorption of moisture from atmosphere. The FSBC/CS blends were prepared manually. The blending weight ratio of CS / (FSBC + CS) are 0%, 10%, 30%, 50%, 70%, 90% and 100%. The pyrolysis experiments were carried out at 600 °C and different blending ratios and also performed at CS blending ratio 70% and different temperatures. The solid heat carrier used in the study was high-temperature boiler ash fetched from a 75 t/h CFB boiler in a power plant. The composition of the boiler ash was analyzed by an X-ray fluorescence spectrometer Analyzer (S4 PIONEER Supplement, Bruker AXS). Table 2 shows some physical properties of the boiler ash.

2.2. Apparatus and procedure

As shown in Fig. 1, the experimental setup consists of six essential sections. These sections are heat carrier feeder ($\Phi 60 \times 6$ mm, 500 mm length), coal/biomass feeder ($\Phi 50 \times 6$ mm, 200 mm length), pyrolyzer ($\Phi 100 \times 6$ mm, 400 mm length), quenching tank ($\Phi 140 \times 6$ mm, 120 mm length), cooling system and a temperature controller. In the upper part of the pyrolyzer, there is a mixer which makes the solid heat carrier and the coal/biomass mix fully to guarantee a rapid heat transfer between them. In order to make the condensable gas fully cooled, the cooling system consists of three condensers in series. The first is cooled by running water, and the second and third are further cooled by ice water placed in a stainless steel container. The temperature controller is connected by K-type thermocouples to the heat carrier feeder and pyrolyzer. The entire system is well sealed.

For every experimental run, 1500 g of boiler ash was loaded into the heat carrier feeder, then was heated by the electrical heater to a set temperature, such as 800 °C, 750 °C or 700 °C, which made the

material pyrolyze at different temperature. The pyrolyzer was heated by the electrical heater to about 400 °C to make up for heat losses. Then 100 g of coal/biomass or their blends was loaded into the coal/biomass feeder. Before experiments pure nitrogen was introduced into the whole setup to ensure air removed. After the heat carrier was heated to the desired temperature, the ash and the coal/biomass particles were dropped into the pyrolyzer at the same time by opening the valves under the two feeders. To prevent the loss of pyrolysis gases, the valves were instantly closed once the ash and the coal/biomass particles fell into the pyrolyzer. The ash and the coal/biomass particles were mixed sufficiently by gathering and scattering many times with the action of the internals in the mixer at the upper part of the pyrolyzer, which lasted a few seconds. At the same time, the coal/biomass particles were pyrolyzed preliminarily during the process of mixing. Then the mixed particles fell into the under part of the pyrolyzer and were pyrolyzed further. The volatile products flowed into the cooling system where the condensable volatiles were collected. The non-condensable gas at ambient temperature kept on flowing into a gas bag to collect for subsequent analysis. Experiments lasted for at least 20 min, until no further significant release of gas was observed. After the completion of pyrolysis, the residual solid blends of ash and char were discharged into the quenching tank to cool to the ambient environment and then were discharged to weigh and recorded. The solid char yield was calculated by the following formula: weight of char = $A - (1500 - B)$, where A is the weight of the discharged blends of char and boiler ash, B is the weight of the leftover boiler ash in the heat carrier feeder. The gas was analyzed by the gas analyzer then to convert weight yield. The yield of liquid product was obtained by calculating weight increase of the cooling tubes which were weighed before test. In this study, each test was carried out at least two times to ensure the repeatability.

2.3. Products analysis method

The chemical compositions of uncondensable gas from pyrolysis of the coal/biomass were analyzed by gas analyzers. Three types of gas analyzers were used to determine the composition of the gaseous product. The permanent gases, such as H₂, CH₄ and CO were analyzed with a SP-2305 gas chromatograph equipped with a TCD detector (5A molecular sieve column and pure argon as carrier gas). The hydrocarbon gases of C₂–C₄ were analyzed with a GC-1790 gas chromatography equipped with a FID detector (C₁₈ column and pure nitrogen as carrier gas). The content of CO₂ in the gas was analyzed by an Orsat gas analyzer.

The liquid products included tar and water. Firstly, the liquid products were poured out from the condensers into a separating funnel where the liquid was separated into tar and water after setting at least 10 min. The tar remained in the condensers was rinsed by tetrahydrofuran (THF). Then THF was removed by a rotary ZFQ-85A evaporator at the conditions of 70 °C, atmospheric pressure and 50 rpm. The two parts of tar were blended together and then were extracted by n-hexane with an ultrasonic SB 25–12 DTDN extractor under the conditions of 40 kHz, 500 W, 25 °C, and 30 min. After extraction, the tar was separated into light-oil (n-hexane soluble) and asphaltene (n-hexane insoluble). Fig. 2 shows the analysis procedure of the liquid product.

The characteristics of the solid char were analyzed including ultimate analysis, ash melting point and composition of the char-ash. Ultimate analysis of the char for carbon, hydrogen, nitrogen, and sulfur were carried out with a CHNS/O Elementary Analyzer (Vario Micro cube, Germany). The ash melting point was analyzed by an Ash Melting Point Analyzer (5E-AF-3, China). The composition of ash was analyzed by an XRF Analyzer (S4 PIONEER Supplement, Bruker AXS).

In addition, to determine the possible synergetic effect during the co-pyrolysis process of FG/CS blends, the measured values were compared with the calculated ones from the additive Eq. (1) below.

Table 1
Proximate and ultimate analyses (wt.%) of Fugu subbituminous coal and Corn stalk.

Sample	Proximate analysis (ad)			HHV (MJ/kg)	Ultimate analysis (dry)					H/C
	M	A	V		C	H	N	S	O ^a	
FSBC	4.82	3.20	36.35	29.91	77.29	4.36	0.97	0.16	14.02	0.67
CS	3.54	5.50	73.94	15.75	45.97	5.59	0.70	0.23	42.01	1.46

ad: air dried base; dry: dried base; M: Moisture; A: Ash; V: Volatiles.

^a By difference.

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