Laboratory and pilot scale studies on fast pyrolysis of corn stover

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Abstract: In order to design a hot ceramic ball heated down-flow tuber reactor for the pyrolysis liquefaction of agricultural residues, the devolatilization characteristics of pulverized corn stover were studied in a Laminar Entrained Flow Furnace (LEFF) with the temperature range of 800–950 K. The flow characteristics of corn stover particles and ceramic balls in a vertical square tube were measured and analyzed by the techniques of particle image velocimetry (PIV). The devolatilization experiments were carried out in an isothermal manner and the maximum of volatile matter of pulverized corn stover particles was 72.68% with the residence time of 189 ms at 950 K. Parameters of the first order Arrhenius equation for describing the pulverized corn stover flash pyrolysis were determined. In the PIV experiments, the corn stover particle axial velocity were measured at different cross-sections of the tube for different conditions. For the tuber connected with a suction pump, the maximum velocity was about 0.79 m/s, determined by the flow rate of the suction pump. Finally, a hot ceramic ball heated down-flow tuber reactor was constructed for pilot demonstration. The capacity of the reactor was 110.5 kg/h of corn stover powder and the yield of bio-oil was 41.6% of the feed stock.

Keywords: Corn stover, pyrolysis, bio-oil, particle image velocimetry
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1 Introduction

Before 1980s, the agricultural residues were combusted directly for cooking and heating for households in the rural areas in China. The energy efficiency was as low as 10% for this kind of application. There were nearly 700 million tons of agricultural residues produced annually in China, 60% of which were used as fuel by direct combustion and 20% as feedstock for industrial conversion and other 20 % as fertilizers.

Agricultural residues are renewable and carbon dioxide neutral feedstock. The Chinese government has paid great attention to the development and utilization of biomass (i.e. agricultural residues) as an energy resource, and has funded some study of advanced biomass conversion technologies, such as pyrolysis, integrated gasification combined cycle generation, etc, through the national programs, such as the Key Science and Technology Projects, the National Hi-Tech R & D Program.

In recent years, new process and equipment have been developed to liquefy biomass. The fast pyrolysis technology is considered to be a promising and applicable technique to convert biomass into liquid product, the bio-oil.

The knowledge of biomass devolatilization characteristics can help researchers design biomass pyrolysis system more effectively. For the reactions of biomass pyrolysis are under very high heating rates, the research apparatus should be provided with the similar conditions at the heating rate of 1000 K/s or higher[1,2]. Particle Image Velocimetry (PIV) is a whole-flow-field technique providing instantaneous velocity vector measurements in a cross-section of a flow. The PIV technique has been widely used in multi-phase flow studies recently. S. Fohanno et al[3] investigated the effect of collisions on the gravitational motion of large particles in a vertical duct. Kaoru Miyazaki et al[4] studied the particle motion in a gas-solid two-phase spiral flow in a horizontal tube. Shi Huixian et al[5] and Wang Qinhui et al[6] studied the flow field in a circulating fluidized bed in cold state. Wei Mingshan et al[7] studied the flow fields in

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an electrostatic cyclone at different positions with different inlet air velocities. The PIV technique has also been applied to gas-solid two-phase flow study in a horizontal entrained flow bed by Yi Weiming and Li Zhihe, et al[8]. Zhang Dongdong et al[9] and Yang Rengang et al[10] studied the two-dimensional velocity field of particles in gas-solid two-phase jet flow field.

In general, reaction temperature, heating rate and residence time of biomass particles are the three main parameters for the design of pyrolysis reactor. Depending on the type of reactor, biomass particle velocity determines the residence time. In previous studies[11, 12], the authors have developed a down flow tube reactor for the pyrolysis of biomass in which ceramic balls were used as the heat carrier. The reactor is made of steel tube (57 mm in inner diameter) with insulation. During the pyrolysis, biomass particles and ceramic balls were fed into the reactor and the pyrolyzed vapors were discharged to the quench system to produce bio-oil under slightly negative pressure environment generated by a vortex pump. The length of the reactor was determined mainly by trial and error, since the movement and interaction of the biomass particles and ceramic balls were very difficult to be measured at high temperature. With the aid of PIV technique, the movement of particles in a transparent tube could be tracked and measured in a cold state model with Reynolds number as the similitude criterion.

The objectives of this work were 1) to study the devolatilization characteristics of pulverized corn stover in a laminar entrained flow furnace; 2) to study the flow characteristics of corn stover particles and ceramic balls in a vertical square tube by the techniques of particle image velocimetry (PIV); and 3) to design and construct a hot ceramic ball heated down-flow tuber reactor.

2 Materials and methods

2.1 Laboratory-scale pyrolysis apparatus

The raw materials were corn stalk, which was collected from Weifang, Shandong Province. The above ground part without cobs was collected. The corn was the variety of Taidan 119 and stored in the shelter. It was crushed by a high-speed rotary mill, and then screened in a series of sieves. After sieving, the corn stover particles with the size fraction of 0.117～0.173 mm was collected and weighted. The pulverized corn stover particles were then dried at 105℃ for 24 hours.

The experimental apparatus used was a redesigned Laminar Entrained Flow Furnace (LEFF) based on traditional structure of LEFF. The design of the LEFF is shown in Figure 1. Corn stover particles (0.117～0.173 mm) were carried with Argon through a water-cooled feeding tube (cold finger) into a hot stream of plasma heated Argon flowing downwards through a vertical furnace tube at a Reynolds number low enough to ensure the laminar flow. The furnace tube was held at the same temperature as the plasma heated Argon with a silicon carbide electric heater. The corn stover particles were heated rapidly to the Argon flow temperature and decomposed as they flow through the furnace. Because the flow was laminar, the decomposing particles traveled in a narrow stream along the axis of the furnace and could be aspirated into a water-cooled quench collector tube where the decomposition was quenched. The amount of corn stover particles decomposition depends both on the transition time (residence time) between the cold finger and quench unit and on the temperature of the Argon flow in the furnace. The residence time varied over the range of 108～224 ms by changing the displacement between the cold finger and quench unit. The temperatures of Argon flow and furnace tube can be adjusted from 700 K to 1100 K.

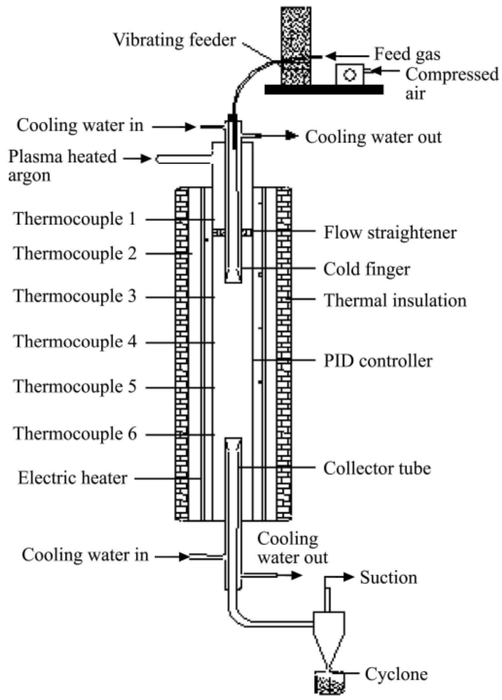

Figure 1 Schematic diagram of plasma heated laminar entrained flow furnace

The furnace consists of a silicon carbide furnace tube (50 mm i.d. inner diameter and 900 mm long) surrounded over the decomposition zone by silicon carbide electric heaters. The displacement between cold finger and quench unit varies from 150～400 mm. Electric heating power is 2 kW. Plasma power is 1.5～50 kW. Argon flow rate is 1.5～2.5 m^3/h. Cold finger corn stover powder feeding rate is about 0.8 g/min. The small flow rates in cold finger are from 0.5 L/min to 1.5 L/min.

2.2 PIV measurement experimental apparatus

The PIV measurement experimental apparatus consists of a vertically mounted square tube, a vibrating feeder, a particle receiver, a vacuum pump and an air pressure stabilizer, etc. (see Figure 2). The vibrating feeder consists of two chute hoppers, hopper angle adjusting bolts, a pneumatic vibrator and an air compressor. The upper chute hopper is used to feed the ceramic balls and the lower chute hopper is used to feed the corn stover particles.

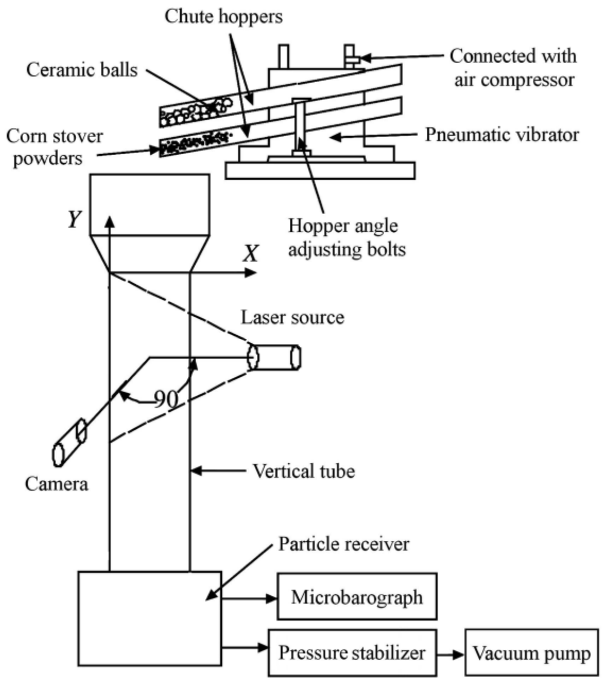

Figure 2 Schematic diagram of PIV measurement system

The PIV system was developed by Beijing Lifangtiandi Science and Technology Development Co. Ltd. It consists of a laser source (Nd: YAG), a personal computer (Dell Dimension 3000) with an image processing system and a CCD camera (AF NIKKOR) with focal length of 50 mm, maximum aperture of f1.4 and resolution of 1600×1200 dpi. The displacement between the camera and the measured flow field is 1040 mm and the displacement between the laser and the measured flow field is 700 mm.

The arrangement of the PIV measurement system is shown in Figure 2. The tube used for this experiment is 1600 mm in length with cross section of 60 mm×60 mm and is vertically mounted on the top of a particle receiver. The light sheet is perpendicular to the side wall of the tube and passes through the axis of the tube and the camera is normal to it. The coordinating origin is selected at the intersection point of left side wall and the inlet of the tube on the plane of the light sheet and the directions of X-axis and Y- axis are shown on Figure 2.

The experimental materials used for this study are corn stover particles (size ranging from 80 to 100 meshes, 0.117～0.173 mm) and ceramic balls (1.0～2.0 mm in diameter). The density of corn stover particles is 114 kg/m^3, and the density of the ceramic balls is 2200 kg/m^3. A vacuum pump and a pressure stabilizer are connected to the particle receiver at the bottom of the tube. For this study, the pressure is 4.0 Pa below the atmospheric pressure when the flow rate of the vacuum pump is 9 m^3/h.

2.3 Hot ceramic ball heated down-flow tuber pilot reactor

A pilot plant of ceramic ball heated down-flow tuber reactor was developed in Shandong University of Technology. The project was supported by National Hi-Tech R & D Program (2001-2005) of China (863 Program). Figure 3 shows the schematic and the photo of the system for the 863 Program. The system consisted of a fluidized biomass combustor, a heater, a down flow tuber reactor, a biomass feeder, a heat carrier separator, a heat carrier recirculating system, a quench unit, etc. Tuber reactor was arranged like letter Z, 45º to the vertical line, to enhance the mixing and heat transfer between the ceramic balls and biomass, and to minimize the thermal stress of the reactor.

Corn stover powder milled with agricultural hammer mill was used as the feed stock for the pyrolysis. Water content of the corn stover powder was 11%-15%. Rice husks were used as feed stock for the combustor to produce high temperature flue gas (1173-1373 K) for heating the heat carrier—ceramic balls (3mm in diameter). Ceramic balls were heated up to 823K in the heater and then fed into the tuber reactor to pyrolyze the corn stover powder at the ceramic ball to biomass ratio of 10:1 by weight. After the pyrolysis reaction, ceramic balls were separated and recirculated to the heater for continuous operation, char was collected and the pyrolysis vapor was pumped to the quench system and condensed into liquid—bio-oil.

3 Results and discussion

3.1 Devolatilization characteristics of pulverized corn stover at high heating rates

The changes of volatile matter (VM) yield with the residence time at different flow temperatures were shown in Table 1.

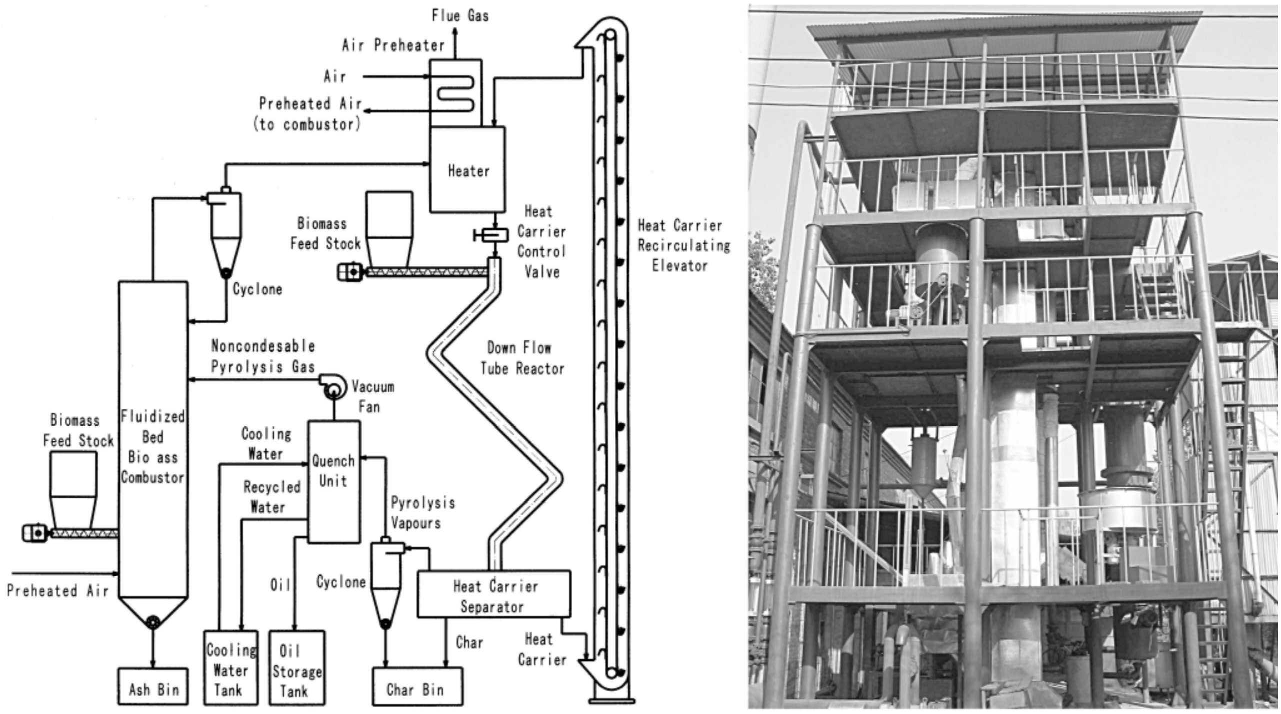

Figure 3 Pilot plant of ceramic ball heated down-flow tuber reactor for fast pyrolysis of corn stover to produce bio-oil funded by 863 Program

Table 1 Yield of volatile mater (%) vs. residence time at different flow temperatures

Flow temperature in LEFF at 800K				
Residence time/ms	128	160	192	224
Percent of volatile/% (yield of VM/%)	44.73	51.13	54.69	59.66
Flow temperature in LEFF at 850 K				
Residence time/ms	121	152	182	212
Percent of volatile/%	51.44	57.20	64.33	66.60
Flow temperature in LFF at 900 K				
Residence time/ms	114	142	170	199
Percent of volatile/%	63.07	64.73	68.59	69.85
Flow temperature in LEFF at 950 K				
Residence time/ms	108	135	162	189
Percent of volatile/%	65.48	66.48	71.37	72.68

Assume that the first order Arrhenius form equation can be used to describe the pulverized corn stover flash pyrolysis.

$$\frac{dW}{dt} = A \cdot (W_\infty - W) \cdot \exp\left(\frac{-E}{RT}\right)$$

Where, W is percent of corn stover powder volatile; t is residence time; W_∞ is final percent of corn stover powder volatile (is 80%); A is apparent frequency factor; E is apparent active energy; R is gas constant; T is particle temperature.

Former researchers have studied the heating history of particle moving in LEFF[13]. The results show that within 5 cm entering LEFF, the particle temperature arises to the gas flow temperature. So we assume the particle temperature is the same as flow temperature in LEFF.

Based on the experimental data, A and E/R can be conducted. So the governing equation of pulverized corn stover is

$$\frac{dW}{dt} = 1039 \cdot (80 - W) \cdot \exp\left(\frac{-4078}{T}\right)$$

The volatilization characteristics of agricultural residues at high heating rates could be investigated successfully in the LEFF. From the results of the experiments, it can be concluded that the yields of volatile pyrolysis products depended both on the final pyrolysis temperature and residence time. Kinetic parameters were obtained from the experimental data. Using the obtained kinetic parameters, a first order kinetic model was used to predict the pyrolytic conversion of corn stover. It is shown from Figure 4 that the theoretical prediction of corn stover pyrolysis is in good agreement with the experimental data.

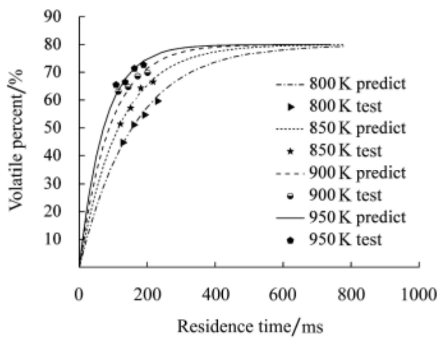

Figure 4 Theoretical predictions and experimental data

3.2 PIV measurement of flow regime in a vertical reaction tube

Figure 5 shows the mixed particle flow field. The concentration of ceramic balls is much lower than that of the corn stover particles. The velocity nephogram of the corn stover particles at different cross-sections of tube is shown in Figure 6 in which the streamlines represent the directions of the resultant velocities. The nephogram shows that the change of the velocity is larger in the region within 0%-20% of the tube width (about 12 mm) from the tube walls, but smaller in central region and the curved streamlines in the same region also exhibit that there are horizontal components of the velocities.

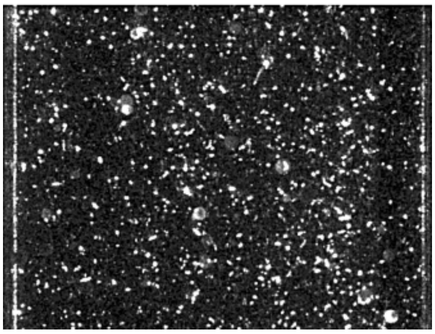

Figure 5 Flow field of the mixed particles of ceramic balls and corn stover particles

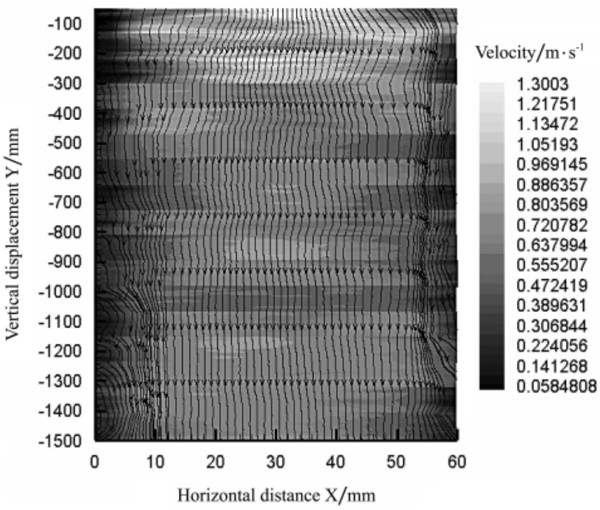

Figure 6 Axial velocity nephogram at different cross-sections of the tube

The corn stover particle axial velocity distributions at different cross-sections of the tube for the mixed flow under suction are shown in Figure 7 and the corn stover particle axial velocity distributions at different cross-sections of the tube for the corn stover particle flow under suction are shown in Figure 8. The X-axis is expressed as the unitary value of horizontal displacements that is the displacement divided by the width of the tube. Figures 7 and 8 show that the corn stover particle axial velocity distributions at various positions for the mixed flow are tub-like curves, whereas, the corn stover particle axial velocity distributions at various positions for the corn stover particle flow are parabola-like curves. The maximum velocities for the two flows are similar (about 0.79 m/s).

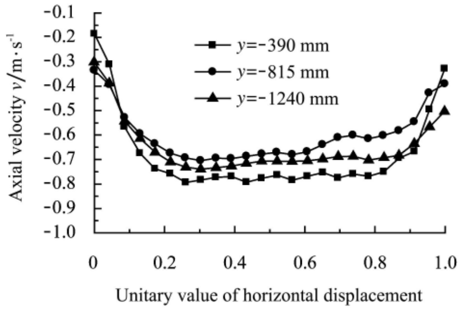

Figure 7 Corn stover particle axial velocity distributions at different cross-sections of the tube for the mixed flow under suction

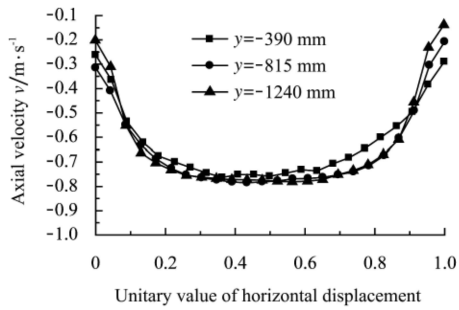

Figure 8 Corn stover particle axial velocity distributions at different cross-sections of the tube for the corn particle flow under suction

The corn stover particle axial velocity distributions at different cross-sections of the tube for the mixed flow with open end are shown in Figure 9 and the corn stover particle axial velocity distributions at different cross-sections of the tube for the mixed flow with open end are shown in Figure 10.

For the mixed flow, the maximum corn stover particle axial velocity with open end condition is about 1.8 m/s (see Figure 9) which is twice more than that under suction condition (0.79 m/s, see Figure 7). This can be due to the air carrying effect caused by the falling of ceramic balls, because the maximum corn stover particle axial velocity for the corn stover particle flow under open end condition is 0.86 m/s (see Figure 10), only slightly higher than that under suction condition. However, this does not happen for the under suction condition in which the maximum corn stover particle velocities for the two flows are about the same, which indicates that for a closed system, the presence of falling ceramic balls has little effect on the maximum corn particle velocity. Perhaps, the average air velocity (gas velocity may be more accurate) in the tube, which is determined by the flow rate of the suction pump, is a dominant factor that affects the corn stover particle velocity. In this study, since the suction pump flow rate is 0.9 m^3/h, the calculated average air velocity in the tube is 0. 694 m/s which agrees well with the velocity distribution of corn stover particles under suction condition (see Figures 7 and 8). Since the density of the ceramic balls is very high (2200 kg/m^3), its motions in all the experimental conditions are the same and exhibit an accelerated motion by the gravity.

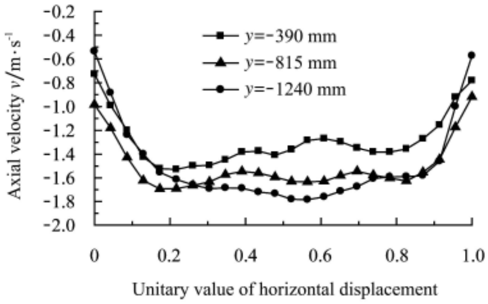

Figure 9 Corn stover particle axial velocity distributions at different cross-sections of the tube for the mixed flow with free end

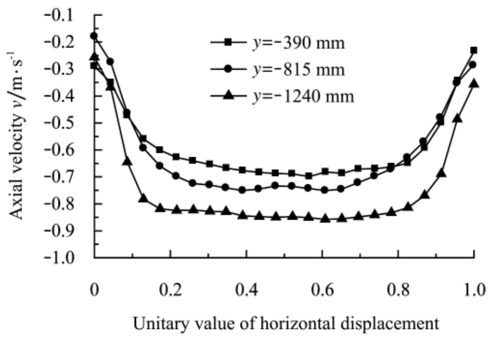

Figure 10 Corn stover particle axial velocity distributions at different cross-sections of the tube for the corn particle flow with free end

3.3 Performance of hot ceramic ball heated down-flow tuber pilot reactor

A series of experiments were conducted on the pilot plant of ceramic ball heated down-flow tuber reactor. For stable operation, the capacity of the reactor was 110.5 kg/h of corn stover powder and the yield of bio-oil was 41.6% of the feedstock. Since the system had a biomass combustion unit for supplying the heat for the pyrolysis, the system was an energy self-sustaining system.

In China, agricultural residues are the most abounding resources of biomass for pyrolysis and characterized by the low density of energy, numerous varieties and scattering supply. The cost of feed stock collecting is the function of collecting radius. If the collecting radius is too big, the cost can be increased greatly. Therefore, the most suitable (economical) operation mode (system) for applying the pyrolysis technology should be a centralized bio-oil refinery combined with several scattered pyrolysis factories for bio-crude production. Biomass feed stocks are first collected and converted into bio-crude in each pyrolysis factory with the collecting radius of 5 km. At this stage, with the increase of densities from about 0.2 of the feed stock to about 1.2 of the bio-oil, the cost for transportation could be reduced significantly. The bio-crude produced in each pyrolysis factory is then transported to the centralized bio-oil refinery and upgraded into bio-oil and other valuable chemicals (products).

The annual biomass yield is around 40000 tones for the area with one pyrolysis factory (5 km in radius). Considering other applications, such as for animal feed, about 15000 tones could be used as the feed stock for bio-crude production. Thus, the daily process rate for each factory must be 50 tones of biomass and 20 tones of bio-crude, counted on 300 working days per year. The equipment capacity is about two tones of biomass per hour, which is fairly large considering the running and maintenance.

A centralized bio-oil refinery should be built in a county (or several counties) for bio-crude upgrading and the bio-crude process capacity should be around 500 tones per day. On this scale, the bio-oil production system could be operated economically.

4 Conclusions

The maximum volatile matter of pulverized corn stover particles was 72.68% with the residence time of 189 ms at 950 K. The first order Arrhenius equation for describing the pulverized corn stover flash pyrolysis was determined. Corn stover particle axial velocity distributions at different cross-sections of the tube for different conditions were measured. For the tuber connected with a suction pump, the maximum velocity was about 0.79 m/s which are determined by the flow rate of the suction pump. Finally, a hot ceramic ball heated down-flow tuber pilot reactor was constructed for pilot demonstration. The capacity of the reactor was 110.5 kg/h of corn stover powder and the yield of bio-oil was 41.6% of the feedstock.

Acknowledgments

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