



Research article

Design and operation of a down-tube reactor demonstration plant for biomass fast pyrolysis



Zhihe Li, Ning Li, Weiming Yi *, Peng Fu *, Yongjun Li, Xueyuan Bai

Shandong University of Technology, Shandong Research Center of Engineering & Technology for Clean Energy, Zibo 255000, China

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ABSTRACT

A novel down-tube reactor demonstration plant has been designed and built with the biomass throughput capacity of over $300 \text{ kg} \cdot \text{h}^{-1}$. The main components of the pyrolysis demonstration plant were described and their performance was tested and evaluated. Preliminary experiments were conducted at the selected range of 400–550 °C to investigate the effect of temperature on the products. The components of non-condensable gases and the chemical compositions of bio-oil were studied. The good features of the reactor (high heat transfer rate, short residence time, good liquidity of biomass and ceramic balls, etc.) were beneficial to increase the yield of bio-oil. The maximum bio-oil yield was obtained at 500 °C up to 52.5 wt.%, with the yield of gas and char being 27% and 20.5%, respectively. The major components of non-condensable gas were CO_2 and CO which were accounted for 90.4 vol.% at 400 °C. The chemical compositions of bio-oil are also affected by temperature above all the proportion of ketones, acids, furans, phenols and sugars. Acids is the main product and the maximum yield is up to 32.3 wt.% at 550 °C. This study has provided a referenced basis for the industrial biomass pyrolytic liquefaction demonstration plant.

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1. Introduction

The serious situation of environmental pollution associated with excessive use of fossil fuels, has drawn attention to an alternative renewable resource. Biomass-based energy provides environmental benefits, economic development and an essential complement for developing countries [1]. Biomass is considered as CO_2 neutral fuel, especially in the form of agricultural or agro-industrial wastes and residues, because biomass growth accumulates amount of CO_2 , via photosynthesis, which is equal to the amount of CO_2 released during its thermochemical conversion. Furthermore, compared with other renewable energy sources, a distinguished merit of biomass is its “materiality”. It is the only carbon resource among the renewable energy sources that can be transformed into liquid fuels [2].

Thermochemical conversion is commonly regarded as the most potential technology route, which transforms lignocellulosic biomass into storable and transportable biofuels and valuable chemicals. Biomass pyrolysis is one of thermochemical processes which is referred as “Thermocracking” and it is a complex process that rapid thermal decomposition of macromolecular compounds occurs in the absence of oxygen to produce liquid, gas and char. The proportion of three products seriously depends on the pyrolysis technique and reaction parameters [3]. In general, biomass fast pyrolysis favors the production of liquid fuel at moderate temperatures (around 500 °C), with high heating

rates ($103\text{--}104 \text{ Ks}^{-1}$), short vapor residence times ($<1 \text{ s}$), rapid char removal from the reaction environment and quick volatile quenching. The highest yield of the main product, bio-oil, is up to 80% on dry-feed basis, given that secondary cracking reaction is minimized [4,5].

Bio-oil, also referred as pyrolysis liquid, pyrolytic oil or bio-crude oil, is the main product of pyrolysis [6,7] and it can be used in diesel engine through emulsification technology [8], in heating and electricity generation or chemicals preparation through physical and chemical methods in current refinery units [9,10], where through the use of suitable catalysts they can be converted into olefins [11], aromatics [12] or hydrogen [13,14]. In addition, biomass pyrolysis also produces bio-char for carbon bio-sequestration and soil conditioning [15].

The reactor is the heart of fast pyrolysis process, which determines the reaction rate and the quality of bio-oil. The commercial fast pyrolysis process generally comprises of biomass reception and storage, feed drying and grinding, pyrolysis conversion, production collection, transport and upgrading. So many reactors have been developed in the lab and large scales [16]. A. V. Bridgwater [5] summarized the demonstration or industrial plants such as bubbling fluid bed, circulating fluid bed, transported bed, rotating cone, ablative, auger and vacuum moving bed reactors. A concise review on different types of reactors is outlined below.

1.1. Fluidized bed reactor

Fluidized bed reactor is previously used in the petrochemical industry for over fifty years. The bubbling fluidized bed reactor is widely

* Corresponding authors.

E-mail address: fupengsdut@163.com (P. Fu).

adopted for biomass fast pyrolysis due to its simple construction, operation and high efficiency of heat transfer. By regulating the flow rate of carrier gas, residence time of vapor can be controlled in a desired range of 0.5 s to 2 s [17]. The high yield of bio-oil is up to 70–75% [5]. However, because of high operation cost and scale-up limitation of heat transfer to bed, there are several larger scale systems operating around the world. The company of Union Fenosa built a 200 kg·h⁻¹ pilot unit in Spain based on the university of Waterloo's fluid bed technology [18]; Dynamotive built the largest plant in Canada (based on the design of RTI) [19], which has been running at a capacity of 8000 kg·h⁻¹ since 2008 [20]; In Biomass Engineering Ltd., UK and Agritherm Canada, 200 kg·h⁻¹ plant are developed respectively (based on the Adelaide University's fluid bed technology in Australia) [21]; Anhui University of Science and Technology in China built a demonstration plant, up to 600 kg·h⁻¹ [22].

1.2. Circulating fluid bed (CFB) and transported bed

A circulating fluid bed (CFB) and transported bed reactor is similar to fluidized bed reactor in many features, except that the residence time of char is the same as for the vapors. The sand heated by the char combustion in the secondary combustor is cycled between pyrolysis units and combustor. The heat and mass transfer is enhanced due to the charred surface of particles abraded during phases colliding. The system has the potential to be scaled up for large throughputs. But the thermodynamics are complex mainly including conduction and convection, heat transfer is not proved at large scales. A circulating fluid bed (CFB) and transported bed reactor system was developed by Metso working with UPM in Finland (based on the Guangzhou Inst's technology), which has been run at a capacity of 400 kg·h⁻¹ [23]. And a large scale facility, Ensyn unit in Canada, was performed by Chemical Process Engineering Research Institute in Greece, which capacity is up to 4000 kg·h⁻¹ since 2007 [24].

1.3. Vacuum moving bed

Vacuum moving bed technology was developed by the university of Laval and Pyrovac in Québec, Canada. Although the heat transfer rate is lower than other reactors, it generates similar liquid products as to the short residence time [17,25]. This reactor can use larger particles than fluidized bed and circulating fluidized in the range of 20–50 mm. However, slow heating rate also results in the low liquid yield, up to 45% on dry feed [4], and the investment and maintenance cost of the design features is high. All above increase the difficulty of the reactor scale-up. Pyrovac built a 3500 kg·h⁻¹ demonstration plant for bark residues [26].

1.4. Auger or screw

Fluidized bed reactor and auger reactor are two common reactors used for biomass pyrolysis [27]. In the process of moving, biomass was mixed with heat carriers (such as sand) by screw system and accomplished the pyrolysis reaction. In comparison, the operation is less complex and the cost is low. However, the liquid yield is lower than fluidized bed because the residence time of vapor is longer (>5 s) and it contacts with the product char. Abritech built a 2083 kg·h⁻¹ plant in Canada which was invented by Auburn University in USA; Lurgi LR built a 500 kg·h⁻¹ plant which was designed by KIT (FZK) in Germany and Renewable Oil Intl developed a 200 kg·h⁻¹ plant based on the technology of Mississippi State University, Michigan State University and Texas A&M University [28].

1.5. Rotating cone reactor

Rotating cone reactor is designed at the University of Twente [29] and developed by BTG [30]. The operation is similar to circulated fluidized bed reactor (transported bed), but the transport of hot sand and

biomass is affected by the centrifugal force rather than carrier gas. The high heating rate and short vapors residence time of the system make it an ideal technology for biomass pyrolysis scale-up work. A 2000 kg·h⁻¹ unit is being operated in Netherlands and USA [5]. A 250 kg·h⁻¹ unit is still operated [9] and a scaled up version of 50 t·d⁻¹ was commissioned in Malaysia in mid 2005 [10].

1.6. Moving bed and fixed bed

The moving bed and fixed bed reactors are usually adopted in the laboratory due to the simple structure. The principle of the process is that an inert carrier gas through a flowmeter is introduced into the reactor which carries vapors to condensers [4,5]. Moving bed and fixed bed reactors were developed by Anhui Yineng Bio-energy Ltd. in China and a 600 kg·h⁻¹ plant was built based on studies at Anadolu University in Turkey and Autònoma de Barcelona University in Spain [23].

1.7. Ablative reactor

The ablative reactor was developed and performed and key work of the process was carried out at NREL (National Renewable Energy Laboratory), University of Nancy, Colorado School of Mines, University of Twente, BBC Engineering Ltd. and Interchem Industries Inc. [31]. Biomass is rapidly heated contacting with hot walls under pressure. Consequently, it is suitable for large biomass particles. A 250 kg·h⁻¹ pilot plant was built by PyTec in Germany, based on the study of Aston U. in UK [32]. A 6 t·d⁻¹ unit has been built in Northern Germany in 2006 and another plant of 50 t·d⁻¹ is being further designed [33].

The down-tube reactor is an alternative free-fall reactor and has been proven to be an adequate reactor for biomass fast pyrolysis. The particles and hot ceramic balls are mixed and flow through the long reaction tube to accomplish the reaction. So there is no inert carrier gas compared to other types of reactors. The cyclic vigorous ceramic balls' movement ensures the heat and mass transfer between phases. Furthermore, the short vapors residence time is attained thanks to the stable negative pressure environment of the internal reactor. The problems related to the process scale-up work have been considered and solved in previous studies [34–36]. Thus, continuous operation can be achieved as char can be continuously removed from reactor and ceramic balls can be recycled in the premise of well-sealed system. Finally, heat supply to ceramic balls and reactor can be achieved by the utilization of biomass furnace.

This paper presents the design, construction and operation of a biomass fast pyrolysis demonstration plant provided with a down-tube reactor, which is based on the technology of biomass directly heated by solid carrier. The aim of this paper is to scale up this original technology developed by the Shandong Research Center of Engineering & Technology for Clean Energy from bench scales to a demonstration plant.

2. Materials and methods

2.1. Experimental set-up

The novel down-tube pyrolysis demonstration plant has been designed and built with the biomass throughput capacity of over 300 kg·h⁻¹. The down-tube reactor is the heart of the pyrolysis system, which is designed based on previous hydrodynamic studies in cold unit and bench-scale units (feed rate respectively 5 kg·h⁻¹, 15 kg·h⁻¹, 50 kg·h⁻¹) [37–39]. Figs. 1 and 2 show the picture and flow chart of the demonstration plant, respectively. It mainly consists of the following parts: the belt conveyor of feedstock, biomass feeding system, flue gas generator, gas-solid heat exchanger, down-tube reactor, ceramic balls and char separator, ceramic balls elevator, char collector, cyclone separator system, bio-oil condensation system (spray tower, demister of pipe bundle condenser, coalescence filters, oil pump, plate heat exchanger).

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