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Inherent process variations between fast pyrolysis technologies: A case study on *Eucalyptus grandis*



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ABSTRACT

Fast pyrolysis of *Eucalyptus grandis* was carried out using different reactor configurations and scale setups, a 0.1 kg/h bubbling fluidised bed (BFB) reactor, a 1 kg/h BFB reactor and a 10 kg/h twin-screw reactor. The influence of lignocellulosic composition, process configuration (reactor configuration, separation and condensation system) and their feed capacity on yields and quality of pyrolysis products was discussed. The increasing reactor scale study had constraints such as the availability of biomass feedstock with inherent changes in the chemical composition and the practicality of the condensation chain. Although the methods employed for phase separation and liquid collection differ from plant to plant limiting the repeatability of results, it was clearly identified that the implementation of efficient solid separation and use of mineral oil as the coolant system were crucial aspects of the mass production of organics. These results suggest that yields and quality of fast pyrolysis at a larger scale are all process-related rather than feedstock conditioned. A comparison of energy balances indicated that fast pyrolysis retained the same energy yield at scales of 1 and 10 kg/h.

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1. Introduction

In light of the current economic climate, the energy crisis and concerns about weather pattern changes, the South African government has taken steps to ensure that South Africa's energy generation is diversified over the next 20 years in such a way as to facilitate climate change mitigation by using among others, biomass as a source of renewable energy [1]. The specie, Eucalyptus grandis, which is used worldwide in Eucalyptus plantations mainly for the paper and pulp industry of South Africa [2], received an increased interest as feedstock for bioenergy production. Rapid growth rates, high wood density and homogeneity of Eucalyptus species constitute real advantages for the implementation of sustainable short-rotation plantings [3] and their thermochemical conversion [4]. Fast pyrolysis has been shown to be an effective method for converting bulky biomass such as wood into valuable and easily transportable products such as bio-oil and char [5], but very little data are available on the pyrolysis of eucalyptus species and specifically *E. grandis*, justifying the need to investigate this topic. Fast pyrolysis is the rapid thermal degradation of biomass in the absence of oxygen, followed by immediate quenching of the product vapours to produce a high yield of liquid product [4,5].

Reactor configurations such as stationary and circulating fluidised bed, ablative, rotating cone, vortex, vacuum and auger reactor and key

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parameters for fast pyrolysis have been extensively reviewed [4,5,7,8, 9], and its main features can therefore be summarised as follows: a moderate reactor temperature of approximately 500 °C; fast removal and rapid cooling of vapours from the heated zone resulting in vapour residence times below 2-3 s; a high particle heating rate within the heated zone; a short solid residence time to minimise catalytic side reactions; and rapid quenching of product vapours to limit secondary reactions and fractionation of bio-oil. The fast pyrolysis of woody biomass has been extensively studied using fluidised bed reactors of various plant scales ranging from 0.15 to 20 kg/h [10-18], but the direct comparison of the reactor scale and configurations used and the differences in the condensation system has not been investigated for a single biomass. Comprehensive reviews demonstrated that fluidised bed reactors, or variations of fluidised bed reactors such as transported bed and circulating bed reactors, are most commonly used for research and industrial applications of biomass fast pyrolysis [4,8]. Fluidised bed reactors are often selected due to their superior mass and heat transfer properties and the efficient mixing of the biomass and heat carrier particles, which can be achieved by fluidisation [19,20]. However, studies conducted at the Karlsruhe Institute of Technology (KIT) in Germany have focused on using an Auger type fast pyrolysis reactor, referred to as the twin-screw reactor [6]. This reactor was chosen because it was already applied on large technical scale for fast pyrolysis of other materials. Unlike fluidised bed reactors where an inert gas stream and a sand bed are used as transport, the twin-screw reactor configuration makes use of mechanical action (rotating parallel screws) to induce mixing of

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the biomass and heat carrier avoiding an inert gas to transport the product vapour and char out of the reactor. Furthermore, seeing that no fluidisation is required for mixing, other heat carriers can be used such as a mixture of steel balls, which can be recycled via an external heater system. The advantages of using steel balls instead of sand as heat carrier have been reported as increased specific biomass throughput in the reactor (10 kg/h with sand and 20 kg/h with steel balls) as well as the elimination of pyrolysis product contamination due to sand entrainment when using sand as the heat transfer medium [6].

Because of its increasing interest as feedstock for the production and upgrading of fast pyrolysis bio-oil to fuel oil qualities, showing high yields [11] and improved thermal properties [21,22], Eucalyptus wood, more specifically one of the 'big nine' species [3], *E. grandis*, has been used in this study. This woody feedstock was converted using two different fast pyrolysis continuous reactors configurations at three different scales to illustrate the impact of the variability of the chemical composition of the lignocellulosic feedstock and the great number of difficulties in interpreting scale-up effects based on product yields and their quality.

2. Material and methods

2.1. Biomass characterisation

The biomass used in this investigation was provided by the Richards Bay Mill of Mondi South Africa, situated on the eastern coastline of South Africa. Two separate batches were used for the experimental work conducted in this study. Batch 1 was used for the experimental work conducted in South Africa, while batch 2 was used for the experimental work performed in Germany at the Karlsruhe Institute of Technology (KIT). Samples from both batches were subjected to lignocellulosic characterisation at the University of Stellenbosch (SU). These samples were milled and sieved using an SM 100 Retsch mill and an AS 200 Retsch shaker, isolating the biomass particle size range from 250 to 425 µm for lignocellulosic characterisation. Lignocellulosic analyses were done in accordance with the TAPPI standard methods for characterising woody biomass (T264 om-88, T211 om-85) and the standard methods described by Browning [23]. Full lignocellulosic characterisation was conducted for batch 1, while the lignocellulosic characterisation of batch 2 was limited to the moisture, ash, extractives and Klason lignin content determination. Elemental analysis of the biomass was done using a TruSpec CHN Vario EL III Leco elemental analyser. Both, lignocellulosic composition of the raw materials and elemental composition of feedstock and pyrolysis products are presented in Tables 1 and 2, respectively.

2.2. Continuous fast pyrolysis plants

A 1 kg/h fast pyrolysis bench scale plant was used for the experimental work conducted at Stellenbosch University, while two devices, a 0.1 kg/h laboratory plant and a 10 kg/h Process Demonstration Unit (PDU), were used for the experimental work conducted at KIT. Duplicate experimental runs were conducted at 500 °C with the biomass feeding rate set to the design specifications of each reactor configuration. The biomass used for each reactor configuration

Table 1

Lignocellulosic composition of Eucalyptus grandis.

	Batch 1 [24]	Batch 2	[12]
Moisture (wt.%, ar)	7.9 ± 0.3	9.4-10.1	7.6
Ash (at \pm 550 °C) wt.%, db	0.5 ± 0.1	0.5	0.4
Extractives wt.%, db	2.6 ± 0.2	4.1 ± 0.4	-
Lignin wt.%, db	15.2 ± 1.5	26.5 ± 2.0	27
Holocellulose wt.%, db	85.1 ± 2.4	$68.9\pm2.4^{\rm b}$	61 ^b

^aBy difference; ^bCorrected for biomass water content.

was dried before use and had an initial moisture content of less than 10 wt.% (Table 1).

2.2.1. Process description of the FP_{0.1 kg/h} plant

The 0.1 kg/h fast pyrolysis (FP_{0.1 kg/h}) plant at KIT is typically used for initial pyrolysis testing before conducting experimental work on a larger scale. Fig. 1 gives a schematic representation of the FP_{0.1 kg/h} plant.

Inside the furnace, a single cyclone separator followed the 4 cm (internal diameter) bubbling fluidised bed (BFB) reactor. Silica sand was used as heat transfer medium in the reactor and nitrogen as the carrier gas. The product vapour exiting the cyclone was first cooled down to room temperature by a single pass, counter-current shell and tube heat exchanger, before passing through two well-insulated condensers at 0 °C (using crushed ice and water). After these first three condensers, the vapour stream was passed through two electrostatic precipitators (ESPs) in series, operated at 8 kV and 4 kV, respectively, and then through a cryogenic trap (kept at -50 °C using dry ice and ethanol). Both bio-oil fractions were recovered and analysed. Finally, the incondensable gas was quantified by a drum type volumetric flow meter (Ritter Trommel-Gaszähler) before it was purged to the venting system. Experimental runs were duplicated at 505 \pm 2 °C with a constant feed rate set point of 0.1 kg/h.

2.2.2. Process description of the FP_{1kg/h} plant

The 1 kg/h fast pyrolysis (FP_{0.1 kg/h}) plant was used at Stellenbosch University (Fig. 2). A 6.6 kW cylindrical furnace housed a 75 mm (internal diameter) stainless steel bubbling fluidised bed (BFB) reactor, followed by two cyclones in series for solids separation. Sand (AFS 45 fused silica, from Consol minerals) was used as the heat carrier inside the reactor. Pyrolysis product vapours left the furnace via a heated pipe (maintained at 400 °C to prevent undesired intermediate condensation) before entering the cooling tower. The condensation chain consisted of a spray-cooling tower followed by two electrostatic precipitators in series set to 14 kV and 12 kV, respectively. An iso-paraffinic hydrocarbon liquid (Isopar purchased from Engen Petroleum Limited) was used as direct contact quenching medium in the cooling tower. A monitoring system recorded all measured process temperatures, pressures and flow rates. A detailed description of the experimental setup has been given by Carrier *et al.* [24].

The process results in one single phase, homogeneous bio-oil, most of which was collected in the liquid collection vessel below the cooling tower during each run. The bio-oil was separated from the mineral oil using a funnel. The amount of bio-oil left in the condensation chain was determined by washing the condensation chain with technical grade acetone, recovering the wash liquid and weighing the residue after the acetone had evaporated from the wash liquid. The char product was recovered from both char pots and subsequently analysed. Experimental runs were duplicated at 500 °C with a constant feed rate set point of 0.85 kg/h.

2.2.3. Process description of the FP_{10kg/h} plant

The 10 kg/h process demonstration unit ($FP_{10 \ kg/h}$) using the twinscrew reactor (Fig. 3) has been described by Henrich and Dahmen [6, 25]. The $FP_{10 \ kg/h}$ was designed to use steel balls (1.5–2 mm) as the heat transfer medium in order to increase the overall heat transfer coefficient, which is typically ranging from 10 to 60 Wm⁻¹ K⁻¹ for a screw type reactor [26]. Milled biomass (<5 mm) was introduced at the left side of the twin-screw reactor, while the hot steel balls (550 °C) of ca. 1.5 mm diameter were introduced at an offset as shown in Fig. 3.

The reactor was operated at a temperature set point of 500 °C. The steel balls and biomass were mixed by the revolving twin screws (120 rpm). All the over-size char particles and steel balls fell down from the reactor and were recycled via the heated bucket elevator system at 600 °C. Product vapours and char were entrained from the reactor with a moderate nitrogen stream to the first condenser (C₁), a single pass shell and tube heat exchanger equipped with an automated

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