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Full Length Article

Biomass fast pyrolysis in a drop tube reactor for bio oil production: Experiments and modeling

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HIGHLIGHTS

- Experiments on bio-oil production via biomass fast pyrolysis in a drop tube reactor (DTR).
- Satisfactory mass balances lying between 88 and 104% for different temperatures and particle sizes.
- The reactor temperature and particle size were found to have a major influence on the pyrolysis products distribution.
- The pyrolysis experiments are supported by pyrolysis products characterization.
- Understanding keys on the biomass fast pyrolysis in a DTR are given and discussed in the light of the modeling results.

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ABSTRACT

Woody biomass fast pyrolysis in Entrained Flow Reactor (EFR) is studied both with experiments in a lab-scale drop tube reactor (DTR) and simulations with a 1-D model. The parameters of the study are temperature (450–600 °C), woody biomass particle size (370–640 μm) and gas residence time (12.6–20.6 s). The most critical phenomena affecting the bio-oil yield are considered in the model: heating of the biomass particles, slip velocity of the biomass particles varying with biomass/char properties, biomass pyrolysis and tar cracking. The analyses of all products – char, bio-oil and gas – also brought information on the advancement of the pyrolysis and cracking for the different tests. The reactor temperature and particle size were found to have a major influence on the pyrolysis product distribution. The production of bio-oil reaches a maximum of 62.4 wt.% at 500 °C for the 370 μm particles. The particle conversion advancement is then estimated at 94% at the reactor exit. The bio-oil yield is lower at higher temperatures for a constant particle size due to tar cracking. At 550 °C, increasing the particle size from 370 μm to 640 μm induces a decrease of the bio-oil yield from 48.3 to 34.8 wt.%, which was shown to be due to incomplete pyrolysis of the particles, because of a too short residence time as well as a too long heating time of particles. The pyrolysis conditions – temperature, particle size – were not found to have any significant influence on the bio-oil properties, such as acidity.

1. Introduction

Actual data as well as serious forecasts on the decreasing availability of fossil resources and the increasing pollution in the future, state on the necessity of more sustainable policies in the different fields of energy, agriculture, industry and services [1]. Biomass, if managed in a sustainable way, represents a renewable energy resource. Woody biomass was used since mankind domesticated fire for various applications such as heating and cooking. Nowadays, these applications remain the major ones for energy recovery from biomass by combustion, especially in the poor countries.

Pyrolysis is another thermochemical way to convert biomass into liquid bio-oil, gas and solid char. The biomass pyrolysis reaction is performed in the absence of oxygen and in a typical temperature range of 450–600 °C. The proportions of gas, solid and liquids highly depend on the pyrolysis process operating conditions [2]. The choice of the technology is conditioned by the nature of the most desired pyrolysis product at the reactor downstream. For instance, fast pyrolysis performed at temperatures of 400–600 °C and at high heating rates, allows maximizing the bio-oil yield. Bio-oil has nearly the same calorific value as the initial wood; it is more easily transportable and has a higher energy density

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Nomenclature

C_d	Drag coefficient, –
C_{p_i}	Mass calorific capacity of i, J/kg.K
d_{es}	Equivalent spherical diameter, m
g	Standard gravity acceleration, m/s^2
h_{pe}	Convective transfer coefficient between particle and gas, W/m^2.K
$\dot{m}_{i,e}$	Mass flowrate of i going from gas environment into the particle, kg/s
m_j	Mass of a pyrolysis reactant/product (j = wood, bio-oil, gas, char, non-condensed water), kg
M_i	Molecular weight of gas species i, kg/mol
$P_{ray,p}$	Radiative power received by the particle, W
P_{sat-H_2O}	Vapor pressure of water at saturation, Pa
r_j	Reaction rate of j at T_p, mol/s
Re_p	Reynolds number for particle, –
R_{ig}	Ideal gas constant, J/(mol·K)
S_p	Particle surface area, m^2
x_i	Volume fraction of gas species i
T_i	Temperature of i, K
$T_{out-cold\ trap}$	Temperature at the exit of the cold trap, K
v_{slip}	Particle slip velocity, m/s
$V_{mol-STP}$	Volume of one mole at STP, m^3/mol
$\dot{V}_{N_2}$	N_2 volumetric inlet flowrate, m^3/s
$\dot{V}_{tot}$	Total gas volumetric flowrate, m^3/s
Y_j	Mass yield of a pyrolysis product (j = non-condensed water, bio-oil, gas, char), wt.%
ΔH_j	Reaction enthalpy of j at T_p, J/mol
Δ_t	Duration of a pyrolysis experimental run, s
ρ_g	Gas density, kg/m^3
ρ_p	Particle density, kg/m^3

(energy per cubic meter) than the raw biomass. Nevertheless, it presents some drawbacks related to its acidic nature, high oxygen and water contents causing problems for storage and combustion [3].

Biomass fast pyrolysis for bio-oil production is most commonly performed in Fluidized Bed Reactors (FBR). These reactors provide a very good heat transfer to the biomass particles by conduction and convection, which allows reaching high heating rates. However, some difficulties are induced by the presence of the heat carrier solid (fluidization, separation from the pyrolysis products etc...) [3]. The Entrained Flow Reactor (EFR) could constitute a simpler alternative to FBR. This type of reactor is well-known for coal gasification and is then operated at high temperature (1500 °C) and high pressure (over 30 bars) to produce a syngas that is rich in H_2 and CO. In such a reactor, solid particles, ground to size under millimeter, are fed by the top. They are submitted to a high heat flux, while they drop along the reactor. The solid residence time is typically of a few seconds in the reactor. The principle of EFR (fine biomass particles dropped in a hot reactor) could be used for fast pyrolysis, by adapting the operation conditions and especially the gas environment and temperature so as to maximize the bio-oil yield.

Some disadvantages of EFR compared to FBR can however be anticipated: lower heat transfer to the biomass particles, need for fine grinding of biomass – energy costly step - to reach a complete conversion before leaving the reactor.

Despite of these, the EFR technology is still very attractive due to its operation simplicity. The number of studies dealing with biomass pyrolysis in FBR is huge, while no more than a few tenths of studies deal with biomass flash pyrolysis in EFR conditions for bio-oil production. To our knowledge, only two attempts were made to develop fast pyrolysis in an EFR at pilot scale [4]. The biomass feeding rate was between 50 and 100 kg/h and in both cases, the reactor was heated by combustion gases of a propane burner [4–7]. One main challenge associated with this technology seemed to lie in the control of the biomass particle residence time, which should be long enough to ensure heating of the whole particle up to devolatilisation temperature [4], and short enough to limit tar cracking [5].

All other studies investigating biomass pyrolysis in EFR conditions were performed in electrically heated drop tube reactors (DTRs) [8–14]. These lab-scale reactors well reproduce some important EFR characteristics such as heat flux, residence time and particle size. The reactor dimensions, operating conditions and main results are given in Table 1. In most of these studies, different parameters were varied (temperature, biomass particle size, biomass feeding rate, inlet gas flowrate) with the objective to determine the optimal values so as to maximize the bio-oil yield. The most appropriate temperature for all studies was between 500 and 600 °C, while there was no common optimal particle size from all considered studies. The pyrolysis temperature should come from a compromise between biomass particle extent and secondary tar cracking [8,12,13], the latter one also depending on vapor residence time. Increasing the particle size induces limitations to heat transfer inside the solid, as well as a shorter solid residence time in the reactor, which may cause incomplete pyrolysis [8]. The solid initial density also has a direct influence on particle residence time [13].

Even if all together, these studies allow identifying the most important phenomena and issues associated with bio-oil production from biomass particles in EFR conditions, none of them has been coupled to a modeling approach which could give a more precise comprehension of the conversion process and help evaluating the respective contributions of the different parameters on the overall result.

On the other hand, a few models have been developed to simulate biomass gasification in EFR. However, most of them mainly focus on the chemistry of pyrolysis [15] without considering the influence of particle size on heat transfer limitation or residence time. This aspect was considered in other models [16,17] validated for higher temperatures representative of gasification process rather than fast pyrolysis to bio-oil one. Gorton et al. [6] developed an interesting model of biomass fast pyrolysis, in parallel with their experiments in the above-mentioned pilot-scale EFR [5,7]. This model provided insight into the phenomena, as well as equations which can be used in the design of industrial reactors, such as simple kinetic laws for biomass pyrolysis and tar cracking.

In the present work, our aim is to improve comprehension of entrained flow fast pyrolysis of biomass using both experimental and modeling approaches. The experiments are performed in a lab-scale drop tube reactor, in conditions representative of EFR operation. The parameters of the study are temperature, woody biomass particle size and gas residence time. For each experimental condition, the solid, gaseous and liquid yields are measured, and characterization of all types of products – gas, bio-oil, solid residue – are performed. The objective is to have a good insight into the influence of tested parameters on all yields and properties, and also to bring basis data to a model developed in parallel to the experiments. The model aims at giving a better comprehension of the