



Kinetics of pyrolysis and combustion of spherical wood particles in a fluidized bed



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ABSTRACT

The kinetics of wood pyrolysis and combustion of residual fuel at different particle diameters and temperatures was investigated. A known mass of wooden spheres was fed at the top of a fluidized bed reactor filled with olivine particles and fluidized with nitrogen. The concentration of H₂, CO₂, CO and CH₄ was on-line monitored with gas analyzers. An irreversible first order reaction was applied to describe the biomass pyrolysis. The rate constant was dependent on the average temperature of wood particle, obtained by solving the transient one-dimensional problem of heat conduction in a sphere. The rate for an irreversible second order reaction between the residual fuel and oxygen at the fluid–solid interface, which takes a finite resistance to mass transfer into account, was adopted to describe the combustion. The semi-empirical kinetic models for pyrolysis and combustion were able to describe, with certain limitations inherent to model simplifications, the experimental transient results of molar flow rates of major released species. A statistical model based on the results of the factorial design of experiments (3²) confirmed a statistical significant effect of temperature and wood particle diameter on the gas yield and time of pyrolysis, respectively.

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1. Introduction

The exponentially growing use of fossil fuels as combustible material has not only drastically increased the cost of petroleum (about 100 \$ per barrel today), but it also has caused serious environmental damages with important economic implications. So, reducing fossil fuels dependence is imperative to face the contemporary challenges in the field of energy generation. In fact, many different aspects of pyrolysis and combustion involving promising alternative biofuels, such as bioethanol [1,2], biobutanol [3,4], and biomass [5,6] have been recently reported in the literature. In the particular case of the later, it essentially occurs because biomass is a cheap renewable energy source that reduces net greenhouse gas emissions (i.e., biomass is carbon neutral, which means there is an equilibrium between the CO₂ emission in the atmosphere and its consumption in photosynthesis) [7].

Biomass can be converted in gas by means of different thermochemical processes. Among these, gasification seems to be the most interesting one [8], particularly if coupled with state-of-

the-art technology, like Integrated Gasification Combined Cycle (IGCC) [9]. Gasification process has been extensively investigated and there are lots of studies that describe in details the involved mechanisms [10,11]. Anyway, it is important to remember the solid fuel is consumed through chemical reactions at high temperature (often ≥ 773 K) to form basically CO, H₂, CH₄ and CO₂. However, sulfur and nitrogen compounds, as well as an undesired liquid mixture of phenols and polycyclic aromatic hydrocarbons (PAH), usually referred to as tar, are also produced. Tar causes many technical problems downstream gasification process, mainly filter clogging and malfunctions to gas turbine [12–14]. A well-established strategy that reduces the tar production, which for practical purpose is also applied to modify the composition of gas phase [10], is the use of fluidized bed gasifiers [15] considered in the current investigation.

Experimental results at equilibrium and equilibrium models are useful to define thermodynamic limits [16,17] in fluidized bed gasifiers. However, they fail to explain many other important aspects of biomass gasification that limits their use for industrial interest. For instance, equilibrium models do not well reproduce results of methane and hydrogen concentrations in gas phase [18,19]. Although recent modified thermodynamic models have contributed for solving some of these drawbacks [20], a better

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comprehension of the involved phenomena requires the investigation of the kinetics of pyrolysis.

Comprehensive and simplified models have been suggested to study the dynamic of pyrolysis [21–36]. Simplified models usually assume that: (i) pyrolysis may be described by a single global first-order reaction with a rate constant dependent on an apparent activation energy [21,22,24,26,32]; or (ii) pyrolysis involves an infinite number of reactions described by a distribution function of activation energy and a single shared pre-exponential factor [24,27,28,30,37]. In summary, the simplified approaches always neglect the complex chemistry of pyrolysis and many times do not consider the transport limitations. Moreover, all these theoretical studies are focused only on particular responses (such as the rate of consumption of biomass and overall conversion from biomass to volatile mass) that usually do not include predictions of the kinetics of released species.

Despite the disadvantages of the many important assumptions in the simplified modeling approaches, the current investigation does not involve a comprehensive pyrolysis model [33,35], which would be more difficult to be solved and would require the knowledge of several physical properties, transport and kinetic parameters. Instead of it, as sometimes occurs in the literature [34,36], the manuscript is focused on a semi-empirical approach. It has the benefit of involving a more reduced number of parameters without neglecting the most important resistances to mass and heat transfer typically observed in simplified approaches [e.g.; 24,27,28,30,37].

A detailed set of kinetic experimental results of formation of major gas species during the pyrolysis of wood (H_2 , CO_2 , CO and CH_4) and combustion of the residual tar/char mixture (CO_2) was also currently obtained at different particle diameters (0.005, 0.01 and 0.015 m) and temperatures (973, 1073 and 1173 K). These data were not only used to estimate experimentally the gas yield and pyrolysis time, but they were also applied to check the reliability of the proposed semi-empirical kinetic models for pyrolysis and combustion. At this point it is important to highlight that a different semi-empirical model was also suggested to describe the stage of combustion of residual tar/char, as well as the formation of CO_2 .

A statistical model based on a factorial design of experiments (3^2), and the already mentioned kinetic pyrolysis model were also applied to verify the effect of the investigated factors (particle diameter and temperature) on responses (gas yield and pyrolysis time) whose practical importance in such a system was extensively presented in the literature [5,34,36].

2. Materials and methods

2.1. Experiments

The experiments were carried out in a bed made of stainless steel (internal diameter = 0.11 m) externally heated by a 6 kW

electric furnace. The bed was partially filled with particles of olivine (equivalent diameter = 4.8×10^{-4} m; particle density = 2500 kg m^{-3}) whose catalytic activity favors the chemical conversion of tar. Such a mineral has also good resistance to mechanical erosion typically found in the fluidized environment [38]. For all the runs the bed was initially heated with air up to the desired operating temperature, so nitrogen was used to fluidize a known mass (see Table 1) of wooden (*Ostrya carpinifolia*) homogeneous spheres fed into the reactor. A probe was used to deliver the biomass just above the bed surface of olivine particles (see Fig. 1). At the end of the pyrolysis step, air was forced through at the bottom of the gasifier in order to burn the residual tar/char mixture. The adopted minimum fluidization velocities [39] and physical chemistry properties of the wood particles [40] were already presented in the literature.

The gas mixture leaving the gasifier always crossed a cyclone and a ceramic filter to separate elutriated solid particles such as fly ashes. Then it was cooled down by means of two stainless steel condensers with cold water as refrigerant, followed by other two glass condensers cooled with a mixture of water and diethylene glycol. A sample of cold gas (≈ 1.3 liters per minute) was pumped into the IR and TCD online analyzers used to determine the volume composition of CO , CO_2 , CH_4 and H_2 . Both these gas analyzers were connected to a PC by means of RS232 port, to monitor the data as a function of time. Several thermocouples, a pressure transducer and two mass flow meters were used to measure the temperature, pressure and flow rates of fluidized gas, respectively. A sketch of the experimental apparatus with the main piping and controls is shown in Fig. 1.

The entire experimental procedure was repeated with three different wood particle diameters (0.005, 0.01 or 0.015 m) at three different temperatures (973 K, 1073 K or 1173 K). It means that a set of 9 (3^2) different operating conditions summarized in Table 1 was currently considered.

2.2. Theoretical considerations

Two single-step chemical mechanisms were suggested to describe separately the process of wood gasification and the combustion of residual tar and char. Based on them, two different semi-empirical approaches that include important simplifications were applied to model the kinetics of pyrolysis (Model I) and combustion (Model II). For instance, external resistances to heat and mass transfer were neglected during the stage of pyrolysis, shrinkage and diffusion effects of formed gas were excluded; the reaction rates were dependent on the average temperature of the particle (i.e.; internal gradients of temperature were not considered), and the combustion of residual tar/char mixture takes place at the fluid–solid interface.

The existence of cracks or structural failures during pyrolysis of wood, which was already clearly evidenced in the literature [34],

Table 1
Operating conditions and design matrix for estimating the main effects of particle diameter (d) and temperature (T) on gas yield (Y) and overall pyrolysis time (τ_{dev}).

Run	$m \times 10^3$ (kg)	Natural variables		Coded variables		Y (%)	τ_{dev} (s)
		d (m)	T (K)	X_1	X_2		
1	2.40	0.005	973	−1	−1	47.2	42
2	3.20	0.01	973	0	−1	46.1	70
3	2.65	0.015	973	+1	−1	46.9	114
4	2.90	0.005	1073	−1	0	62.4	48
5	2.60	0.01	1073	0	0	59.6	61
6	2.80	0.015	1073	+1	0	60.4	95
7	2.70	0.005	1173	−1	+1	63.7	35
8	3.00	0.01	1173	0	+1	68.9	68
9	2.70	0.015	1173	+1	+1	67.6	89

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