



Full Length Article

Devolatilization of millimeter-sized biomass particles at high temperatures and heating rates. Part 2: Modeling and validation for thermally-thin and -thick regimes



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ABSTRACT

This paper analyzes the relevance of intraparticle gradients in the devolatilization of millimeter-sized biomass particles under the high temperatures and heating rates typical of the industrial facilities. Experimental data (intraparticle temperature history, size evolution, devolatilization times and volatile release rates) were obtained in tests with isolated particles 3–15 mm in diameter in a flat flame reactor under controlled combustion conditions. These results were compared with the predictions of two different models that alternatively neglect or consider the internal gradients, after deriving model-dependent kinetics from the data; this allowed drawing conclusions regarding the range of validity of each model. Whereas the thick-particle model reasonably reproduces the behavior of the devolatilizing particles for the whole range of test conditions and particle sizes explored, the thin-particle model is unable to fit the trends observed in the data for different sizes. Neglecting the internal gradients leads to a delayed onset and a more abrupt release of the volatiles, which is relevant e.g. in the simulation of biomass-fired burners. Additionally, the experimental data set also allowed assessing the validity of two common assumptions regarding the char oxidation stage, i.e. oxidation limited by O₂ external diffusion and sequential character of devolatilization and char oxidation; both are likely applicable in the conditions explored.

1. Introduction

When a biomass particle is introduced in a high-temperature atmosphere, it experiences different processes. First, there is an initial heating period and a mass loss related to the moisture content of the particle. Next, the volatile species are released (stage of pyrolysis or devolatilization) and diffuse to the surrounding atmosphere where, if oxygen is present, may burn creating a volatile flame around the particle. Devolatilization leaves a carbonaceous residue (char) which, depending on the environmental conditions, may be subsequently oxidized, remaining finally only the inorganic fraction or ash. Since biomass usually contains large amounts of volatiles, the relative importance of this stage on the overall conversion process of the particles is greater than for other fuels such as coal or coke [1].

The pyrolysis entails the thermal degradation of the organic material of the fuel and the production of a large number of chemical compounds; however, for most engineering applications, they can be classified into three groups: permanent gases, pyrolytic liquid (bio-oil/tar) and char, or even more simply into volatiles and char. From the

early studies performed by Bamford et al. [2], numerous efforts have been devoted to model the combustion of biomass particles and a great variety of models differing in the number of intermediate ‘stages’ considered can be found in the literature. The most elaborated models involve multi-step semi-global schemes which take into account the effect of secondary tar reactions, potentially more relevant at high temperatures and sufficiently long residence times (e.g. [3–5]), whereas other authors assume that the raw fuel decomposes directly to each product according to a series of parallel independent reactions (one-step multi-reaction schemes; e.g. [6,7]). Finally, the simplest approaches (and those most used in commercial computational codes mainly due to the greater computing cost of the others) describe the conversion of the fuel into products by means of one-step global schemes (e.g. [8,9]).

Independently of the complexity of the pyrolysis model, the corresponding kinetic parameters must be carefully determined or selected. Most of the devolatilization kinetics published are derived from thermogravimetric analysis (TGA) at low temperatures and heating rates, and may not be applicable to the much more severe thermal conditions

Abbreviations: FFB, flat flame burner; TNP, thermally-thin particle regime; TKP, thermally-thick particle regime

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Nomenclature		X_{fC}	fixed carbon content of the original biomass particle
A_p	external surface area of the particle (m^2)	X_H	fraction of moisture content of the original biomass particle
A_v	frequency factor for devolatilization (s^{-1})	<i>Greek symbols</i>	
C	char content of the particle (kg)	ε	emmissivity
c_p	specific heat ($J kg^{-1} K^{-1}$)	η	variable of progress for the devolatilization reaction
D_{O_2}	mass diffusivity of oxygen ($m^2 s^{-1}$)	φ	reaction mechanism factor for char oxidation (= 1 if CO ₂ is product; = 2 if CO)
D_p	particle diameter (m)	λ	thermal conductivity ($W m^{-1} K^{-1}$)
E_v	activation energy for devolatilization ($J mol^{-1}$)	σ	Stefan-Boltzmann constant ($5.98 \cdot 10^{-8} W m^{-2} K^{-4}$)
<i>err</i>	error parameter	ρ	density ($kg m^{-3}$)
H	moisture content of the particle (kg)	<i>Subscripts</i>	
H_{evap}	enthalpy of vaporization of water ($J kg^{-1}$)	0	values for initial time
H_v	enthalpy of devolatilization ($J kg^{-1}$)	b	biomass
k_{diff}	specific oxygen diffusion rate coefficient ($kg m^{-2} s^{-1} Pa^{-1}$)	c	char
k_v	devolatilization rate (s^{-1})	d	dried particle
m	mass (kg)	dev	devolatilization
m'_c	mass flow rate of char consumed by heterogeneous oxidation ($kg s^{-1}$)	eff	effective thermal conductivity of the particle
m'_v	mass flow rate of volatiles released during the devolatilization ($kg s^{-1}$)	evap	evaporation
n	parameter describing the spread of a Rosin-Rammler distribution	exp	experiment
Nu	Nusselt number	fl	volatile flame
Pr	Prandtl number	g	free gas stream
P_{O_2}	partial pressure of oxygen (Pa)	j	experimental data
Q	heat flow ($J s^{-1}$)	k	test condition
r	radial coordinate (m)	O ₂	oxygen
Δr	size of the computational cell (m)	oxid	oxidation
R	universal gas constant ($8.314 J mol^{-1} K^{-1}$)	p	particle
Re	Reynolds number	pr-k	predicted value according to thermally-thick particle model
R_p	particle radius (m)	pr-n	predicted value according to thermally-thin particle model
sh	shrinkage factor	time	time component of the error defined to derive the kinetic parameters
t	time (s)	w	wall of the inner surface of the FFB's quartz tube
T	temperature (K)		
v	velocity ($m s^{-1}$)		
V	volatile matter content (kg)		
Vol	volume (m^3)		

prevailing in pulverized-fuel flames [10]. Comparatively, the data obtained from tube furnaces or fluidized bed reactors are much scarcer; Di Blasi [11] reviewed the temperature ranges explored in these tests by several authors and the corresponding one-step kinetics, and found roughly two groups regarding activation energies: $E_v = 60\text{--}90 \text{ kJ mol}^{-1}$ and $E_v = 124\text{--}174 \text{ kJ mol}^{-1}$ obtained at high (up to 1400 K) and low ($< 700\text{--}800 \text{ K}$) temperature, respectively, which illustrates the spread of the data available and the relevance of the experimental conditions on the kinetics derived. Besides, the temporal evolution of the fuel conversion process may be affected by the thermal transient period which can be very different depending on the heating rate; that could greatly explain the differences between E_v 's derived from different conditions.

In what regards the modeling of the heat and mass transfer inside the particles, two main situations can be distinguished, depending on the conditions (ambient temperature, heating rate, size and type of the particles, kinetics of the reactions). In some cases, the heat conduction across the particle is so fast that the internal temperature profile is nearly uniform and the heating, drying and devolatilization occur sequentially; this is usually known as the thermally-thin (particle) regime. In the so-called thermally-thick regime, important intraparticle temperature gradients result from the fact that the internal transfer phenomena are not fast enough and several sub-processes may occur simultaneously, although in different parts of the particle. The models based on the thermally-thin particle assumption are computationally

much cheaper than those forced to resolve the internal transfers inside the particles, and for this reason they are usually implemented in CFD codes as long as they are thought to be valid. In this regard, an *a priori* criterion based on the Biot number (external vs. internal heat transfer rates) may be used to determine the applicability of that assumption [12,13]; there is a region (of conditions), however, in which such a rough criterion is not conclusive, and this seems to be the case of biomass combustion in many facilities (or, within a single facility, in the distribution of sizes used).

There are relatively few studies aimed at experimentally quantifying the importance of the 'thickness' of the biomass particles on its behavior in a combustion facility. Most of the experimental work on this subject could be *a priori* classified into one of the regimes just described according to the Biot number criterion: many dealt with centimeter-sized particles in moderate conditions, which clearly lies in the thermally-thick regime [12,14,15], and less with finely pulverized (say, $< 3 \text{ mm}$) samples in more aggressive environments [16–20], clearly in the thin-particle range. To the authors' knowledge, only the work of Lu et al. [21,22] refers to the combustion of few-millimeter biomass particles at severe conditions; in what regards specifically the characterization of the effect of intraparticle gradients in the predictions of temperature and mass loss evolution [21], those authors used nearly fixed particle sizes ($\sim 10 \text{ mm}$) in their experiments, concluding that the gradients where relevant and should be taken into account in the models. The complete 'few millimeter-range' is, however, of considerable interest,

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