MODELING OF BIOFUEL PELLETS TORREFACTION IN A REALISTIC GEOMETRY

by

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Low temperature pyrolysis also known as torrefaction is considered as a promising pretreatment technology for conversion of biomass into a solid biofuel with enhanced properties in terms of lower moisture and volatile matter content, hydrophobicity and increased heating value. A thermal treatment leads to a non-uniform temperature field and chemical reactions proceeding unevenly within the pellets. However the temperature is assumed to be uniform in the pellets in the majority of models. Here we report on the model of single pellet biomass torrefaction, taking into account the heat transfer and chemical kinetics in the realistic geometry. The evolution of temperature and material density in the non-stationary thermo-chemical process is described by the system of non-linear partial differential equations. The model describing the high-temperature drying of biomass pellet was also introduced. The importance of boundary effects in realistic simulations of biomass pellets torrefaction is underlined in this work.

Key words: *torrefaction, biomass, pellet, heat and mass transfer, kinetics, modeling*

Introduction

Biofuel pellets have been attracting more attention in recent years as a renewable energy source playing an important role in the shift towards more sustainable energy systems. However, they are characterized by relatively low heating value, fast biological degradation and hygroscopic nature. In order to address these issues, there is a need of pretreatment techniques [1-6] to convert pellets to an intermediate solid biofuel, which is more suitable for efficient thermochemical conversion by pyrolysis, gasification, combustion or co-firing with conventional solid fuels.

Torrefaction is a low temperature pyrolysis carried out under inert atmosphere conditions in the temperature range of 200-300 °C for relatively long reaction times and typically at low heating rates (<50 °C/min) [1]. During this process, water and a part of the volatiles are released from the biomass, causing a decrease in mass and an increase in energy density [3]. Due to the significant mass loss and a relatively smaller loss of calorific content, the heating value of processed biomass per mass unit increases significantly in the process.

As torrefaction process has been attracting an increasing interest in recent years, many efforts were put to understand deeper the complex mechanisms of wood particles decomposition and to develop a comprehensive model, capturing its most significant features

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[7]. However, the following simplifying assumptions are commonly applied by different authors [8-10]:

- total volume of particles remains unchanged: no solid shrinkage and cracks,
- no heat transfer by convection and radiation within the solid,
- local thermal equilibrium exists between the solid and the gas phase,
- constant thermo-physical properties (specific heat, thermal conductivity),
- unrestricted outflow of gas species from the solid,
- no secondary reactions,
- no moisture content, and
- particles are represented in a one-dimensional, time-dependent domain.

The kinetics of torrefaction reactions [8-17], the physical and thermochemical characteristics of biomass under different operating conditions [1-4, 18-33] were studied by different authors extensively. However most models neglect the non-uniformity of the temperature field along the radius of pellets while assuming the samples as infinitely long slabs in order to simplify the calculations. This approach allows describing the chemical kinetics and heat transfer with one-dimensional model. A comprehensive one-dimensional (1-D) model accounting for the effects of heat and mass transfer, chemical kinetics and drying was developed by Bates and Ghoniem [8]. According to these authors, the thermochemical submodels presented in [8] depend only on previously determined or measured characteristics, avoiding the use of fitting or tuning parameters and enabling a rigorous energy balance of the process. A drying model was also developed that is claimed to give physically consistent results, smooth implementation, and numerical stability. The significant drawback of all 1-D models is that the non-uniformity of the temperature field and chemical reactions within the large particles are not taken into account as well as real geometry and sizes of the particles undergoing torrefaction.

Perre, *et al.* [34] developed a two-dimensional (2-D) coupled model combining heat and mass transfer and kinetics mechanism for the torrefaction of beech wood boards. The predicted temperature profiles were compared to experimental results for beech boards of 5 × 15 × 25 cm.

A very few three-dimensional (3-D) models exist in the literature. A 3-D model of wood torrefaction at 150-250 °C developed by Kadem, *et al.* [35], accounted for heat transfer and drying, but neglected thermal decomposition kinetics.

The detailed heat transfer analysis coupled with chemical kinetics within a cylindrical biomass pellet exposed to torrefaction is of great importance in order to control the torrefaction process and to achieve high quality of final product. Therefore the main objective of this study is to develop a model of a single woody pellet torrefaction taking into account heat transfer, real shape, size, thermal properties of original pellets and phase transformation and chemical conversions during torrefaction process. A new modeling approach is implemented to describe the high temperature drying of biomass pellets undergoing torrefaction.

Torrefaction process model development

Torrefaction is a complex process involving a set of chemical and physical phenomena, *e. g.* heat and mass transfer, moisture evaporation, decomposition kinetics, chemical reactions, changes in material properties. The understanding of chemical and physical phenomena during biomass torrefaction is of fundamental importance in order to design a reactor.

A pellet of a cylindrical shape (fig. 1) with the radius R, length L and the initial temperature T_0, exposed to torrefaction in a fixed bed reactor with temperature of $T_s > T_0$ is considered in this work.

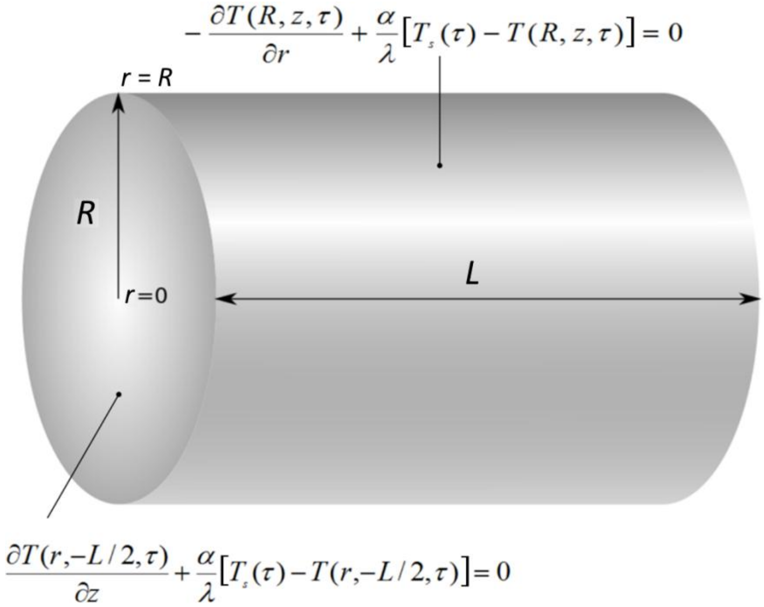

Figure 1. The geometry of pellet exposed to torrefaction

The heat exchange between the bounding surfaces of the cylindrical pellet and the medium is described by Newton's law. It is assumed that the heat transfer within the pellet occurs by conduction, and the heat is produced or consumed due to the chemical reactions and physical changes during the torrefaction. The conservation of the energy for the pellet can be represented by a second order partial differential equation of heat transfer in the cylindrical coordinates r, z:

$$\frac{\partial\{[c_{p1}\rho(r,z,\tau)+c_{p2}\rho_{mc}(r,z,\tau)]T(r,z,\tau)\}}{\partial\tau}=\lambda\nabla^2T(r,z,\tau)+Q(r,z,\tau) \tag{1}$$

$$0\le r\le R,\ -L/2\le z\le L/2 \tag{2}$$

$$Q(r,z,\tau)=\frac{\partial\rho(r,z,\tau)}{\partial\tau}q_1-\frac{\partial\rho_{mc}(r,z,\tau)}{\partial\tau}q_2 \tag{3}$$

The rate of weight loss per unit volume can be expressed in the Arrhenius form as:

$$\frac{\partial\rho(r,z,\tau)}{\partial\tau}=-A_1\left[\rho(r,z,\tau)-\rho_s\right]\exp\left[\frac{E_1}{R_gT(r,z,\tau)}\right] \tag{4}$$

A new modeling approach [36] has been implemented to describe the high temperature drying of biomass pellets undergoing torrefaction. After the initial heating period, once the boiling temperature (100-135 °C [37, 38]) is reached, the drying rate increases in a stepwise fashion. Therefore to describe the rate of the drying process, the following kinetic evaporation model is used, with the reaction factor, $A_2\,[s^{-1}]$ and the width of the drying reaction interval, s [K], chosen to provide evaporation rate, increasing rapidly at the boiling temperature T_b [K]:

$$\frac{\partial \rho_{mc}(r,z,\tau)}{\partial \tau} = -A_2 \rho_{mc}(r,z,\tau)\left[1+\tanh\frac{T(r,z,\tau)-T_b}{s}\right] \tag{5}$$

It is assumed that at time zero the pellet has a uniform temperature T_0 and density ρ_0:

$$T(r,z,0)=T_0 \tag{6}$$

$$\rho\,(r,z,0)=\rho_0 \tag{7}$$

The heat exchange between the pellet surface and the medium is described by Newton's law:

$$-\frac{\partial T(R,z,\tau)}{\partial r}+\frac{\alpha}{\lambda}\left[T_s(\tau)-T(R,z,\tau)\right]=0 \tag{8}$$

$$\frac{\partial T(r,-L/2,\tau)}{\partial z}+\frac{\alpha}{\lambda}\left[T_s(\tau)-T(r,-L/2,\tau)\right]=0 \tag{9}$$

$$-\frac{\partial T(r,L/2,\tau)}{\partial z}+\frac{\alpha}{\lambda}\left[T_s(\tau)-T(r,L/2,\tau)\right]=0 \tag{10}$$

The symmetry conditions can be written as follows:

$$\frac{\partial T(0,z,\tau)}{\partial r}=0 \tag{11}$$

$$\frac{\partial T(r,0,\tau)}{\partial z}=0 \tag{12}$$

The system of partial differential eqs. (1)-(12) is solved using the numerical method of lines, implemented in Wolfram Mathematica software – a powerful tool that has nearly 5,000 built-in functions for computation, modeling, visualization, development, and deployment [39], covering many areas of technical computing.

Results and discussion

We have studied temperature field and density within the beech wood pellet during torrefaction process using eqs. (1-12). The modeling results have been compared to experimental data, obtained by Van der Stelt [10], who performed torrefaction of cylindrical beech and willow particles with diameters between 1 and 2.8 cm and lengths of 10 cm in a fixed bed reactor. The particles were placed in a vertical split tube oven which was electrically heated. Five thermocouples, mounted inside a particle, continuously monitored the temperature at radial positions r = 0, 0.3, 0.7, 1 and 1.3 cm. The pellets and the reactor – initially at ambient temperature – were heated at a rate of approximately 10 °C/min to the final reactor temperature, which was varied between 230 and 300 °C in different experiments [10]. The mass and energy yield of the particle were not reported. The initial moisture content of the particles was reported as approximately 6% [10]. The parameters used in the simulation of torrefaction process are listed in the tab. 1. Some of them were taken from [10], while others were determined by fitting the temperature fields, obtained from calculations, to the experimental results, performed by Van der Stelt.

Table 1. Variables used in the simulation of torrefaction process

No.	Parameter	Symbol	Unit	Value
1	Torrefaction temperature	T_t	K	562
2	Thermal conductivity of wood pellet	λ	$\mathrm{Wm^{-1}K^{-1}}$	0.18
3	Convective heat transfer coefficient	α	$\mathrm{Jm^{-2}K^{-1}}$	18
4	Specific heat capacity of wood pellet	c_{p1}	$\mathrm{Jkg^{-1}K^{-1}}$	1,400
5	Saturated density of wood pellet	ρ_s	$\mathrm{kgm^{-3}}$	420
6	Initial density of wood pellet	ρ_0	$\mathrm{kgm^{-3}}$	700
7	Radius of pellet	R	m	$1.4\cdot10^{-2}$
8	Enthalpy of torrefaction reaction	q_1	$\mathrm{Jkg^{-1}}$	$-6\cdot10^{5}$
9	Activation energy of torrefaction reaction	E_1	$\mathrm{Jmol^{-1}}$	3,500
10	Pre-exponential factor of torrefaction reaction	A_1	$\mathrm{s^{-1}}$	0.6
11	Vaporization heat	q_2	$\mathrm{Jkg^{-1}}$	$1.13\cdot10^{6}$
12	Boiling temperature	T_b	K	420
13	Vaporization reaction factor	A_2	$\mathrm{s^{-1}}$	0.25
14	Width of the drying reaction interval	s	K	10
15	Specific heat capacity of moisture	c_{p2}	$\mathrm{Jkg^{-1}K^{-1}}$	4,200
16	Moisture content	w	%	6

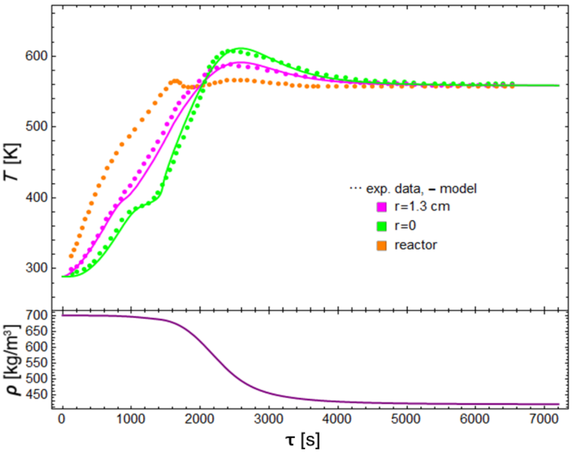

Figure 2. Experimentally measured [10] and modeled temperature profiles at two radial positions *r* = 0, *r* = 1.3 cm at the top plot and density profile at the bottom plot for beech pellets torrefied at 562 K

Experimentally measured [10] and predicted by the model temperature profiles at two radial positions $r = 0$ and $r = 1.3$ cm are presented on the top plot of fig. 2 for the beech cylindrical wood particles, heated up to torrefaction temperature of 562 K (289 °C). The bottom plot illustrates the change in the density of beech wood pellet.

The initial heating periods for both radial positions $r = 0$ and $r = 13$ mm are in a good agreement with the modeling results. The plateaus, corresponding to the drying process can be observed, and are becoming more pronounced towards the centerline of the cylindrical particle as the temperature distribution is non-uniform due to a finite relatively large pellet size, and the higher temperature at the surface of the pellet, $r = R$, leads to faster moisture evaporation there. The model and experimental data demonstrate the centerline temperature decrease after reaching the maximum temperature. However, the model predicts the centerline temperature decrease at a rate faster than the experiment. A significant temperature difference across the pellet along with the exponential temperature dependence of reaction rates leads to a non-uniformity of chemical reactions, that must be taken into account in a realistic model. The heat release due to exothermic reactions resulting in the thermal overshoot is observed after 2,000 s.

Many existing models address the case of long pellets in which the length is much larger than the radius. In this case the boundary effects at the ends of cylinder are negligible compared to the bulk, and the description based on two-dimensional model is sufficient. However, in real applications pellet diameter could be comparable to their length, and therefore three-dimensional model is required [40]. Figures 3a and 3b show how uniform temperature distribution in pellets of finite length changes during torrefaction. Two dimensions of pellets were considered in this work:

– with the length L = 14 mm and the radius R = 14 mm, and
– with the length L = 50 mm and the radius R = 14 mm.

Isotherms close to the cylinder surface gathered more and more closely together since the surface layers are heated faster than the layers close to the pellet center. It can be seen from fig. 3a and 3b that for the pellets of the same radius, R = 14 mm, the temperatures reach higher values and the boundary effects are more pronounced for the shorter pellets with the length L = 14 mm than for L = 50 mm ones.

It can be observed from these simulations that the edges of the pellet burn first, and then heat propagates to the center. This is also confirmed by considering the density profiles. Isotherms shown in fig. 3a and 3b are very far from straight lines along the cylinder axis that should be observed for an infinite cylinder. These results emphasize the importance of boundary effects adequate treatment in torrefaction process simulations of the cylindrical pellets.

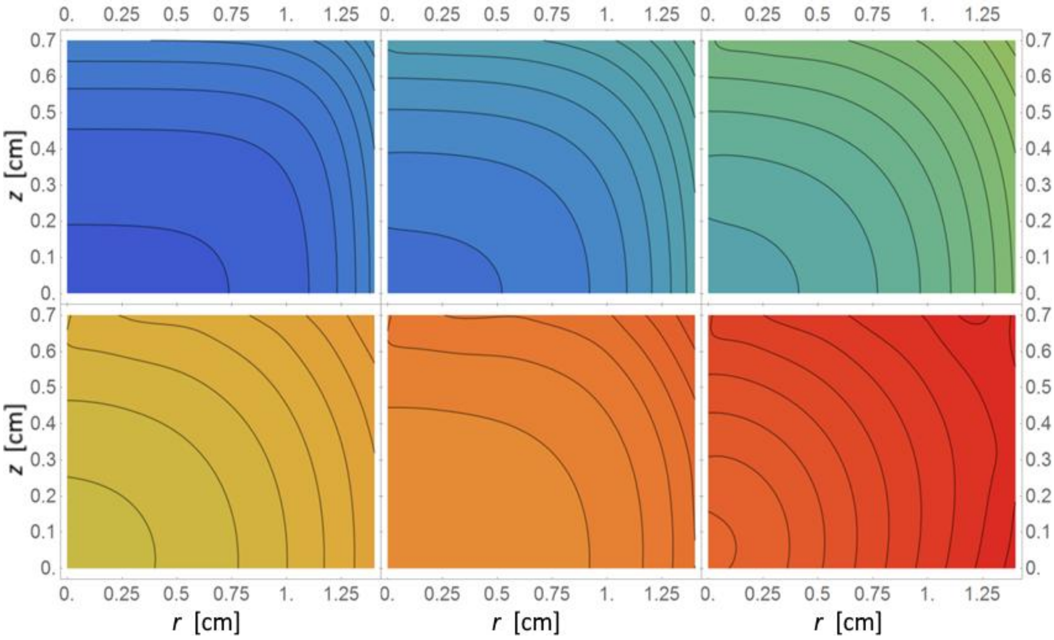

Figure 3(a). Contour plots showing temperature in Kelvin as a function of radial and axial coordinates at different times: 50, 100, 200, 500, 750, 1,000 s (from left to the right) in the pellet of *R* = 14 mm and *L* = 14 mm

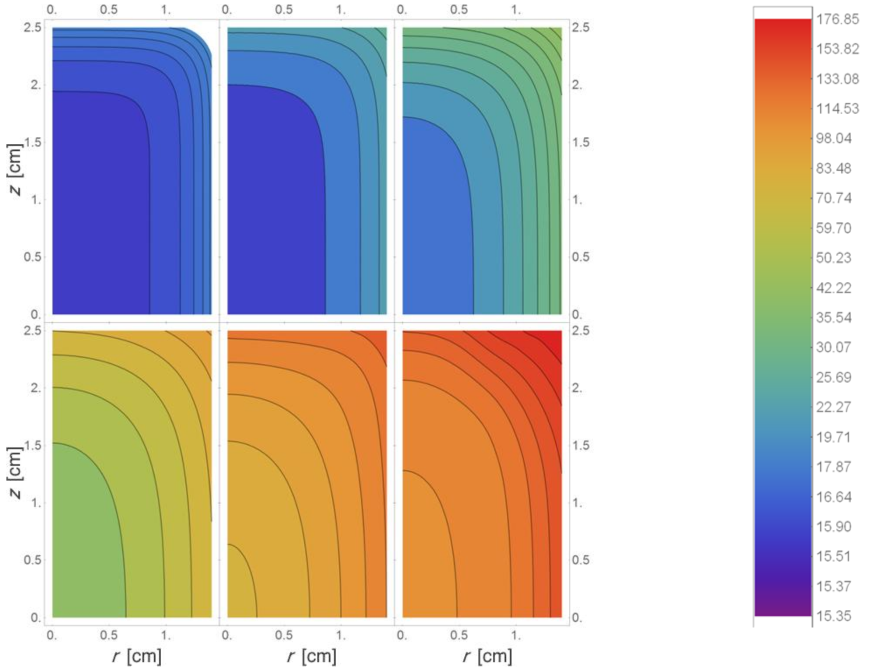

Figure 3(b). Contour plots showing temperature in Kelvin as a function of radial and axial coordinates at different times: 50, 100, 200, 500, 750, 1,000 s (from left to the right) in the pellet of *R* = 14 mm and *L* = 50 mm

Conclusions

This paper presents a model that couples thermal decomposition kinetics with the heat transfer phenomena in a cylindrical pellet exposed to torrefaction. A non-uniform temperature field within the pellets leads to chemical reactions proceeding unevenly. The evolution of temperature and material density in the non-stationary thermo-chemical process was described by the system of non-linear partial differential equations. A high-temperature drying model, describing the drying rate increase in a step-wise fashion, was also introduced. The modeling results are in good agreement with Van der Stelt's experimental data. Two dimensions of pellets were considered in this work:

- with the length *L* = 14 mm and the radius *R* = 14 mm, and
- with the length *L* = 50 mm and the radius *R* = 14 mm.

For the pellets of the same radius, *R* = 14 mm, the temperatures reach higher values and the boundary effects are more pronounced for the shorter *L* = 14 mm pellets than for *L* = 50 mm ones. For pellets of finite size isotherms are very far from straight lines along the cylinder axis that should be observed for an infinite cylinder. These results emphasize the importance of boundary effects adequate treatment in torrefaction process simulations of a cylindrical pellet.

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Nomenclature

A_1	– pre-exponential factor, [s^{-1}]
A_2	– vaporization reaction factor, [s^{-1}]
c_{p1}	– specific heat of wood pellet, [$Jkg^{-1}K^{-1}$]
c_{p2}	– specific heat of moisture, [$Jkg^{-1}K^{-1}$]
E_1	– activation energy, [$Jmol^{-1}$]
L	– length of the pellet, [m]
Q	– reaction heat term, [$Jm^{-3}s^{-1}$]
q_1	– enthalpy of torrefaction reaction, [Jkg^{-1}]
q_2	– vaporization heat, [Jkg^{-1}]
R	– radius of the pellet, [m]
r	– radial co-ordinate, [m]
$\mathrm{R_g}$	– universal gas constant, [$Jmol^{-1}K^{-1}$]
s	– width of the drying reaction interval, [K]
T	– temperature in a pellet, [K]
T_0	– initial temperature, [K]
T_b	– boiling temperature, [K]
T_s	– temperature in the reactor, [K]
z	– axial coordinate, [m]

Greek symbols

α	– convective heat transfer coefficient, [$Wm^{-2}K^{-1}$]
λ	– thermal conductivity of wood pellet, [$Wm^{-1}K^{-1}$]
ρ	– wood pellet density, [kgm^{-3}]
ρ_0	– initial density of wood pellet, [kgm^{-3}]
ρ_{mc}	– moisture density, [kgm^{-3}]
ρ_s	– saturated density of wood pellet, [kgm^{-3}]
τ	– time, [s]
∇^2	– Laplace operator, [m^{-2}]

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