

Torrefaction of Biomass Pellets: Modeling of the Process in a Fixed Bed Reactor

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Abstract—Torrefaction of biomass pellets is considered as a useful pretreatment technology in order to convert them into a high quality solid biofuel that is more suitable for pyrolysis, gasification, combustion, and co-firing applications. In the course of torrefaction, the temperature varies across the pellet, and therefore chemical reactions proceed unevenly within the pellet. However, the uniformity of the thermal distribution along the pellet is generally assumed. The torrefaction process of a single cylindrical pellet is modeled here, accounting for heat transfer coupled with chemical kinetics. The drying sub-model was also introduced. The non-stationary process of wood pellet decomposition is described by the system of non-linear partial differential equations over the temperature and mass. The model captures well the main features of the experimental data.

Keywords—Torrefaction, biomass pellets, model, heat and mass transfer.

I. INTRODUCTION

THE application of biomass pellets for energy production has been rapidly gaining attention in recent years to address environmental concerns and achieve a sustainable - energy supply. Biomass pellets are subjected to mild pyrolysis, also known as torrefaction, to decrease moisture and volatile matter contents and to enhance their energy density, hydrophobicity, and grindability.

Torrefaction is a thermal pretreatment technology performed in the temperature range of 200-300°C [1] under inert or limited oxygen environment, typically at low heating rates (<50°C), and relatively long residence time (about 1 hour). During this process, water and a part of volatiles are released from biomass, causing a decrease in mass and an increase in energy density [2]. Due to a significant weight loss, the energy content of processed biomass per mass unit increases significantly in the process. Torrefaction combined with pelletisation allows to obtain the torrefied pellets (TOP) with an energy content in the range of 18–23 MJ/kg LHV or 20–24 MJ/kg HHV on dry basis [3], [4]. Due to the breakage of hydroxyl groups in torrefaction process, the material loses its tendency to absorb water, so it remains stable and hydrophobic during the storage [1], [2].

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. However, the following simplifying assumptions are commonly applied by different authors [5]-[7]:

- total volume of particles remains unchanged: no shrinkage and cracks of the solid;
- 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;
- negligible initial moisture content;
- particles are represented in a one-dimensional, time-dependent domain.

Most models neglect the non-uniformity of the temperature field and chemical reactions 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 model accounting for the effects of heat and mass transfer, chemical kinetics and drying was developed by [5]. According to these authors, the thermochemical sub-models 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. The significant drawback of all one-dimensional (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 a real geometry and sizes of the particles undergoing torrefaction, causing a significant discrepancy with experimental data.

The detailed heat transfer analysis coupled with a chemical kinetics within a biomass pellet exposed to torrefaction is of pressing interest to enable control of torrefaction process and to achieve high quality of final product. The objective of this paper is to develop a model of single pellet torrefaction accounting for the heat and mass transfer, the non-uniformity of the temperature field as well as chemical reactions proceeding unevenly along the radius of the pellet.

II. TORREFACTION 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

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chemical and physical phenomena during biomass torrefaction is of fundamental importance in order to design a reactor.

A torrefaction model coupling the chemical kinetics, drying and heat transfer is developed in this work to describe the torrefaction of a single wood pellet. The following phenomena and aspects are taken into account in this work:

- heat and mass transfer within the pellet;
- thermal decomposition kinetics;
- moisture evaporation;
- realistic geometry of the pellet;
- non-uniformity of the temperature field and chemical reactions;
- realistic heating protocol.

Torrefaction of a single wood pellet of the radius R , length L and the initial temperature T_0 in a fixed bed reactor at temperature $T_s > T_0$ is considered in this paper. For long pellets ($L \gg R$) we ignore the boundary effects on the sides for simplicity, modeling pellets by infinitely long circular cylinders. It is assumed that the heat transfer within the pellet occurs by conduction and the heat Q is produced or consumed due to chemical reactions and phase changes. The conservation of energy is expressed by a second order partial differential equation of heat transfer:

$$c_p \frac{\partial[\rho(r, \tau)T(r, \tau)]}{\partial \tau} = \lambda \left(\frac{\partial^2 T(r, \tau)}{\partial r^2} + \frac{1}{r} \frac{\partial T(r, \tau)}{\partial r} \right) + Q(r, \tau) \quad (1)$$

$$\tau \geq 0, 0 \leq r \leq R \quad (2)$$

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

where; Q – reaction heat term, J; q_1 – enthalpy of torrefaction reaction, J/kg; q_2 – vaporization heat, J/kg; c_p – specific heat capacity, J/(kg·K); ρ – density of the pellet, kg/m³; ρ_{mc} – density of the moisture, kg/m³; r – radial coordinate, m; τ – time, s.

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

$$\frac{\partial \rho(r, \tau)}{\partial \tau} = -A_1 (\rho(r, \tau) - \rho_f) \exp \left(\frac{E_1}{R_g T(r, \tau)} \right) \quad (4)$$

where ρ_f – final density of torrefied pellet, kg/m³; A_1 – pre-exponential factor, 1/s; E_1 – activation energy, J/mol; R_g – universal gas constant, J/(mol·K).

The following approaches are typically used to estimate the evaporation rate of biomass particles exposed to high temperature drying, pyrolysis, or combustion [5]:

1. first order kinetic evaporation model [8]–[13];
2. equilibrium model [14]–[17];
3. constant temperature model, also known as the heat sink model [18]–[23].

The first order kinetic evaporation model describes the evaporation rate with a first order Arrhenius equation, where the activation energy and pre-exponential factor are chosen to

provide an evaporation rate, increasing rapidly at the boiling temperature [9], [24]. The advantages of this method are that it is implemented easily as an additional chemical reaction in particle models and results in smooth intraparticle temperature gradients [25]. The disadvantages are that some part of moisture is evaporated before the boiling temperature is actually reached, and the kinetic parameters in the literature demonstrate a wide scatter [26].

The equilibrium approach accounts for heat and mass transfer limitations and assumes that the bound and free liquid moisture exist in equilibrium with the local gas phase. The partial pressure of water vapor is fixed by the local saturation pressure. The disadvantage of the equilibrium approach is that it is known to result in depressions in pressure in locations ahead of the drying front and requires simultaneous solution of algebraic and differential equations [25].

The heat sink approach assumes that drying is controlled entirely by heat transfer. When a particle layer has reached the boiling temperature, all energy flowing into that layer is consumed entirely by evaporation of water until drying ceases. It is implemented in models in one of two ways. Either the drying front is assumed to be infinitely thin and the particle is divided into moist and dry layers or the governing equation must include a conditional statement. The former method, though easily implemented, is not necessarily valid in cases where the drying front has a thickness comparable to the particle [13]. The latter method requires special numerical treatment and is not computationally efficient due to the sharp discontinuities it introduces into the governing equation [25], [26].

A new modeling approach has been implemented to describe the high temperature drying of biomass particles undergoing torrefaction [27]. After the initial heating period, once the moisture boiling temperature (100–135°C [18], [28]) is reached, the evaporation rate increases in a step-wise fashion. Hence, to describe the rate of the drying process, the following kinetic evaporation model is used, with the reaction factor, A_2 (1/s) and the width of the reaction interval, s (K), chosen to provide evaporation rate, increasing significantly at the boiling temperature T_b (K):

$$\frac{\partial \rho_{mc}(r, \tau)}{\partial \tau} = -A_2 \rho_{mc}(r, \tau) \left(1 + \tanh \frac{T(r, \tau) - T_b}{m} \right) \quad (5)$$

The local dry basis moisture fraction is given by

$$w = \frac{\rho_{mc}(\tau, r)}{\rho(r, \tau)} \quad (6)$$

It is assumed that at time $\tau = 0$ the pellet has a uniform temperature T_0 and density ρ_0 :

$$T(r, z, 0) = T_0 \quad (7)$$

$$\rho(r, z, 0) = \rho_0 \quad (8)$$

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

$$-\frac{\partial T(R, \tau)}{\partial r} + \frac{h}{\lambda} (T_s(\tau) - T(R, \tau)) = 0 \quad (9)$$

where R – radius of the pellet, m; $T_s(\tau)$ – the temperature of the surrounding medium, K; h – convection heat transfer coefficient between the pellet surface and the medium.

Due to the symmetry requirements;

$$\frac{\partial T(0, \tau)}{\partial r} = 0 \quad (10)$$

The system of Partial Differential Equations (1)-(10) is solved using the numerical method of lines implemented in Wolfram Mathematica (R) software.

III. RESULTS AND DISCUSSION

The modeling results have been compared to experimental data, obtained by [7], who performed torrefaction of

cylindrical beech and willow particles with diameters between 1 and 2.8 cm and lengths of 10 cm. The biomass particles were torrefied in a fixed bed reactor placed in a Carbolite vertical split tube oven that was electrically heated. Argon was used as inert gas to keep the system in absence of oxygen and remove the formed products to the coldtrap. Five thermocouples continuously monitored the temperature inside the particle at radial positions $r = 0; 3; 7; 10$ and 13 mm. The particles 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 [7]. The mass and energy yield of the particle were not reported. The initial moisture content of the particles was reported as approximately 6% [7].

The temperature profiles at two radial positions $r = 0$ and $r = 13$ mm measured experimentally and predicted by the model are presented in Fig. 1 for the beech cylindrical wood particles, heated up to torrefaction temperature of 562 K (289 °C) during 1 hour.

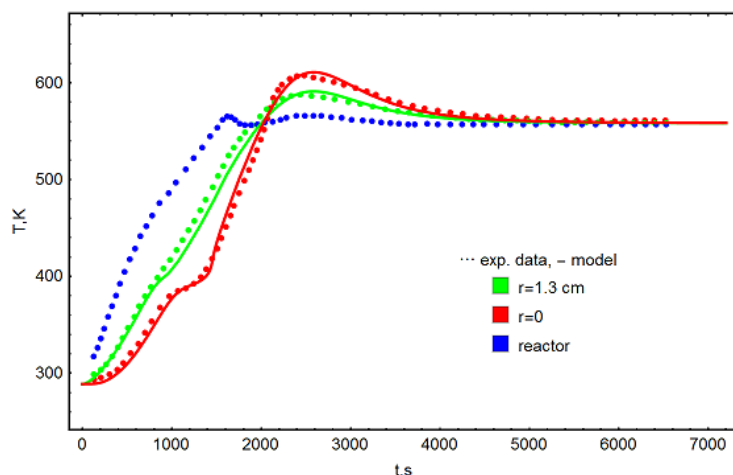


Fig. 1 The experimentally measured [7] and predicted by model temperature profiles at two radial positions $r = 0$ and $r = 13$ mm for beech pellets torrefied at 562 K during 1 hour

The plateaus, corresponding to the drying process, can be observed, and are becoming more pronounced towards the centerline of the cylindrical particle. This phenomenon can be explained by the fact that 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 plateau temperature is higher than the boiling temperature of water.

A significant temperature difference across the pellet, observed for the residence time of 40 min, along with the exponential temperature dependence of reaction rates leads to a dramatic non-uniformity of chemical reactions, that must be taken into account in a realistic model.

The thermal overshoot observed after 2000 s is due to exothermic reactions.

It can be seen from Fig. 1 that the developed model captures all the main features of the torrefaction process, e.g. plateaus corresponding to drying process and the temperature overshoot due to exothermal character of reaction.

The model slightly underestimates the temperature near the pellet surface, while the agreement improves towards the centerline of the pellet. This could be due to a systematic error in the temperature measurement, caused by the finite thickness of the thermocouple, and particularly near the surface of the pellet, where large temperature gradients exist.

This work proves the importance of the non-uniformity of the temperature field and chemical reactions proceeding unevenly for a realistic analysis of heat transfer and chemical kinetics for long cylindrical pellets.

The developed torrefaction model, accounting for the heat and mass transfer, thermal decomposition kinetics and drying, allows to get a deep insight into the process, and will be useful for an optimization of the process and the reactor designing.

IV. CONCLUSIONS

The majority of the models, describing the torrefaction process, assume the uniformity of the temperature field and chemical reactions within the pellets, exposed to torrefaction. This approach allows describing the process by one-dimensional problem of heat and mass transfer, that significantly simplifies the problem solution. We have studied the 2-D torrefaction model for the long pellets, approximating them by infinitely long circular cylinders, and neglecting the boundary effects at the bases. The non-isothermal decomposition of single wood pellet was described by the system of non-linear partial differential equations for the temperature and material density fields. The drying model was also introduced. The modeling results for internal temperature profiles agreed well with the experimental data from the literature.

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REFERENCES

- [1] P. C. A. Bergman, A. R. Boersma, R. W. R. Zwart, J. H. A. Kiel. Torrefaction for biomass co-firing in existing coal-fired power stations. "Biocoal". Technical Report ECN-C-05-013. Petten, The Netherlands: ECN; 2005.
- [2] J. J. Chew, V. Doshi. Recent advances in biomass pretreatment — torrefaction fundamentals and technology. *Renew SustEnerg Rev*, 15 (2011), pp. 4212–4222.
- [3] A. Uslu, A.P.C. Faaij, P.C.A. Bergman. Pre-treatment technologies, and their effect on international bioenergy supply chain logistics. *Techno-economic evaluation of torrefaction, fast pyrolysis and pelletisation. Energy* 33 (2008), pp. 1206–1223.
- [4] P. C. A. Bergman Combined torrefaction and pelletisation: the TOP process, Report No.: ECN-C-05-073, Energy Research Centre of the Netherlands (ECN), Petten, The Netherlands, 2005, pp. 29.
- [5] R. B. Bates, A. F. Ghoniem. Modeling kinetics-transport interactions during biomass torrefaction: The effects of temperature, particle size, and moisture content. *Fuel* 2014; 137: pp. 216–229.
- [6] R. B. Bates. Modeling the coupled effects of heat transfer, thermochemistry, and kinetics during biomass torrefaction. MSc thesis, Massachusetts institute of technology, 2010.
- [7] M. J. C. Van der Sijpe. Chemistry and reaction kinetics of biowaste torrefaction. PhD thesis, Eindhoven University of Technology; 2011.
- [8] W.-C. R. Chan, M. Kelbon, B. B. Krieger. Modelling and experimental verification of physical and chemical processes during pyrolysis of a large biomass particle. *Fuel* 1985;64: pp. 1505–1513.
- [9] C. Di Blasi, S. Branc, Sparano, LaB Manti. Drying characteristics of wood cylinders for conditions pertinent to fixed-bed countercurrent gasification. *Biomass Bioenergy* 2003;25: pp. 45–58.
- [10] W. J. Parker Development of a model for the heat release rate of wood: a status report. Gaithersburg, MD: US National Bureau of Standards; 1985.
- [11] B. Krieger-Brockett, D. Glaister. Wood devolatilization – sensitivity to feed properties and process variables. In: Bridgwater AV, Kuester JL, editors. *Res. Thermochem. Biomass Convers.*. Netherlands: Springer; 1988. p. 127–42.
- [12] S. Shrestha, Cramer S, White R. Time temperature profiles across a lumber section exposed to pyrolytic temperatures. *Fire Mater* 1994;18:211–20.
- [13] K. M. Bryden, K.W. Ragland, C. J. Rutland. Modeling thermally thick pyrolysis of wood. *Biomass Bioenergy* 2002;22:41–53.
- [14] M. G. Grønli A theoretical and experimental study of the thermal degradation of biomass. PhD thesis, Norwegian University of Science and Technology; 1996.
- [15] J. C. Wurzenberger, S. Wallner, H. Raupenstrauch, J. G. Khinast. Thermal conversion of biomass: comprehensive reactor and particle modeling. *AIChE J* 2002;48: pp. 2398–2411.
- [16] H. Lu, W. Robert, G. Peirce, B. Ripa, L. L. Baxter. Comprehensive study of biomass particle combustion. *Energy Fuels* 2008; 22: pp. 2826–2839.
- [17] N. Ouelhazi, G. Arnaud, J.P. Fohr. A two-dimensional study of wood plank drying. The effect of gaseous pressure below boiling point. *Transp Porous Media* 1992; 7: pp. 39–61.
- [18] S. S. Alves, J. L. Figueiredo. A model for pyrolysis of wet wood. *ChemEngSci* 1989; 44: pp. 2861–2869.
- [19] A. Galgano, C. Di Blasi. Modeling the propagation of drying and decomposition fronts in wood. *Combust Flame* 2004; 139: pp. 16–27.
- [20] J. Porteiro, J. L. Míguez, E. Granada, J. C. Moran. Mathematical modelling of the combustion of a single wood particle. *Fuel Process Technol* 2006; 87: pp. 169–75.
- [21] B. Peters, E. Schröder, C. Bruch, T. Nussbaumer. Measurements and particle resolved modelling of heat-up and drying of a packed bed. *Biomass Bioenergy* 2002; 23: pp. 291–306.
- [22] R. Bilbao, J. F. Mastral, J. Ceamanos, M. E. Aldea. Modelling of the pyrolysis of wet wood. *J Anal Appl Pyrolysis* 1996; 36: pp. 81–97.
- [23] J. Saastamoinen, J.-R. Richard. Simultaneous drying and pyrolysis of solid fuel particles. *Combust Flame* 1996; 106: pp. 288–300.
- [24] P. D. Aerts, K. W. Ragland. Pressurized downdraft combustion of woodchips. *Twenty-Third Symp Int Combust* 1991; 23: pp. 1025–32.
- [25] M. Bellais. Modelling of the pyrolysis of large wood particles. PhD thesis, KTH Royal Institute of Technology; 2007.
- [26] B. Peters, C. Bruch. A flexible and stable numerical method for simulating the thermal decomposition of wood particles. *Chemosphere* 2001; 42: pp. 481–90.
- [27] E. Artiukhina, P. Grammelis, G. Sin. Biomass pellet torrefaction: 2D model development, reliability assessment by sensitivity and uncertainty analyses, to be published.
- [28] H. A. Noves, J. I. S. Fernandez-Golfín. Practical evaluation and operation of superheated steam drying process with different softwoods and hardwoods. *Eur J Wood Wood Prod* 1994; 52: pp. 135–138.