NUMERICAL INVESTIGATION OF FLOW REGIMES IN BIOMASS PYROLYSIS

Filipe L. Brandão, filipe_lb@poli.ufrj.br[1]
Gabriel L. Verissimo, gabrielverissimo@poli.ufrj.br[1]
Albino J.K. Leiroz, leiroz@mecanica.ufrj.br[1]
Manuel E.C. Cruz, manuel@mecanica.coppe,ufrj.br[1]

[1]Departamento de Engenharia Mecânica, Universidade Federal do Rio de Janeiro

Abstract. *Pyrolysis involves many physical and chemical phenomena that are complex to model in detail. When pyrolysis occurs in a fluidized bed, the combination of hydrodynamics, mixing/segregation, chemical reactions and heat transfer phenomena hinders the development of models with realistic kinetic mechanisms. Therefore, the utilization of simplified kinetic schemes becomes important for obtaining practical results, mainly for CFD modelling of gasification process. Different models for biomass pyrolysis have been described in the literature. For Euler-Euler CFD studies, the description of the pyrolysis process by kinetic models has been reported as more appropriated in the literature. The present work considers the wood pyrolysis process in a fluidized bed reactor using an Euler-Euler approach. The present comparative study contributes to understanding the differences among the main models, and verifies their applicability for simulating the pyrolysis of wood. Additionally, the influence of employing laminar and turbulent flow assumption over the products composition predicted by the CFD model is also analyzed. It is observed that there are not significant differences in the reactor temperature between laminar and turbulent regimes assumptions. However, it is found that when assuming turbulent regime, the reactor temperature is slightly lower for all the kinetic models studied. Furthermore, it is shown that depending on the pyrolysis kinetic model, the reaction rates profiles along the reactor diverge substantially between laminar and turbulent assumption. For some of those models, the difference is only in the intensity with the reaction rate achieving greater values when turbulent regime is assumed. For another models, it is possible to see clearly that the laminar assumption delays the reaction rates. For the products distribution, it is not reported any significant deviance between values of laminar and turbulent regimes. Among all the kinetic models studied, the greatest relative difference between flow regimes is for char production with the value of 8.7%.*

Keywords*: Pyrolysis, CFD, Euler-Euler, biomass*

1. NOMENCLATURE

$R_{\phi k}$ = source term of the conservation equation for the quantity ϕ_k
$\mathbf{V}_k$ = velocity vector of phase *k*
u_k = internal energy of phase *k*
Y_{ki} = mass fraction of specie *i* of the phase *k*

Greek Symbols
ϕ_k = a scalar or vectorial quantity of phase *k*
ε_k = volumetric fraction of phase *k*

Subscripts
k = phase *k*
i = chemical specie i

Abbreviations
ANC = biomass pyrolysis model proposed by Anca-Couce et al. (2014)
BJR = modification proposed by Blondeau and Jeanmart (2012) of the biomass pyrolysis model proposed by Ranzi et al. (2008)
GMB = biomass pyrolysis model composed of the wood pyrolysis model proposed by Gronli and Melaaen (2000) and kinetic parameters for tar cracking obtained by Boroson et al. (1989)
MB = biomass pyrolysis model proposed by Miller and Bellan (1997)
MFIX = multiphase flow with interphase exchanges
NCG = non-condensable gas
PAR = biomass pyrolysis model proposed by Park et al. (2010)
RAN = biomass pyrolysis model proposed by Ranzi et al. (2008)
SHA = biomass pyrolysis model proposed by Sharma et al. (2014)

2. INTRODUCTION

Pyrolysis involves many physical and chemical phenomena that are complex to model in detail (Souza-Santos, 2004). When pyrolysis occurs in a fluidized bed reactor, the combination of hydrodynamics, mixing/segregation, chemical reactions, and heat transfer phenomena hinders the development of models with realistic kinetic mechanisms. Therefore, the utilization of simplified kinetic schemes becomes important for obtaining practical results (Morf, 2001), especially for CFD modeling of gasification processes (Gómez-Barea and Leckner, 2010). Different models for biomass pyrolysis have been described in the literature (Souza-Santos, 2004; Gómez-Barea and Leckner, 2010; Di Blasi, 2008;

Anca-Couce et al., 2014). For Euler-Euler CFD studies, the description of the pyrolysis process by kinetic models has been reported as more appropriated in the literature (Gómez-Barea and Leckner, 2010). Three formulations of kinetic models are usually employed for pyrolysis description: the global one-step reaction model, the competitive reactions model, and the parallel reactions model (Souza-Santos, 2004; Di Blasi, 2008; Anca-Couce et al., 2014). The global one-step reaction model is a simpler approach to the pyrolysis description considering a single reaction constant. Since the global one-step model is incapable of capturing different char-to-volatile ratios in the pyrolysis product compositions as temperature evolves, it has not been considered in the present work. Parallel and competitive reaction models consider different reaction constants for each pyrolysis product, thus allowing for the temperature effect quantification on the yields of volatile and char. Besides, the parallel reactions model is, in principle, applicable to any biomass, as long as its composition is known in terms of the principal constituents.

The present work considers the wood pyrolysis process in a fluidized bed reactor using an Euler-Euler approach. Results for three specific competitive models for wood pyrolysis, proposed by Gronli and Melaaen (2000), Park et al. (2010) and by Sharma et al. (2014) are compared to the parallel reaction pyrolysis models by Miller and Bellan (1997), Ranzi et al. (2008), Blondeau and Jeanmart (2012) and Anca-Couce et al. (2014). A significant number of works on modeling of biomass (Bradbury et al., 1979; Miller and Bellan, 1997; Morf, 2001; Souza-Santos, 2004; Ranzi et al., 2008) and wood (Boroson et al., 1989; Gronli and Melaaen, 2000; Sharma et al., 2014) pyrolysis is available in the literature. Despite works using CFD approaches being also present in the literature (Xue et al., 2011; Xue et al., 2012; Xiong et al., 2013; Mellin et al., 2014; Sharma et al., 2015), the coupling between pyrolysis and CFD models is considered a relatively new and limited issue (Xue et al., 2011). The present comparative study contributes to understanding the differences among the main models, and verifies their applicability for simulating the pyrolysis of wood. Additionally, the influence of the commonly employed laminar flow assumption (Xue et al., 2011; Xue et al., 2012; Xiong et al., 2013; Mellin et al., 2014; Sharma et al., 2015) over the products composition predicted by the different kinetic models employed in the CFD model is also analyzed.

3. MATHEMATICAL FORMULATION

3.1 Governing equation and closures models in the Euler-Euler approach

The gas-particle flow is modeled using an Euler-Euler approach (Syamlal and Pannala, 2011), in which a manner that differential partial equations are used to describe the governing equations of mass, momentum, internal energy and species for gas and solid phases, which may be represented in a general way as shown in Eq. (1) for a quantity ϕ_k. It must be noted that the subscript k in Eq. (1) denotes the phase of the quantities and the quantity ϕ_k assume the values shown in Eq. (2).

$$\frac{\partial \varepsilon_k \phi_k}{\partial t} + \nabla \cdot (\varepsilon_k \phi_k \mathbf{V}_k) = \nabla \cdot (\Gamma_{\phi_k} \nabla \phi_k) + R_{\phi_k} \tag{1}$$

$$\phi_k = \begin{cases} 1, \text{for continuity equation} \\ \mathbf{V}_k, \text{for momentum conservation equation of phase } k \\ u_k, \text{for internal energy conservation equation of phase } k \\ Y_{ki}, \text{for conservation equation of specie } i \text{ of phase } k \end{cases} \tag{2}$$

As in previous computational studies of fluidization and thermo-chemical (Yu et al., 2007; Gerber et al., 2010; Xue et al., 2012; Xiong et al., 2013; Verissimo et al., 2014) processes, a 2D Cartesian domain is used for the reactor simulation. The turbulence of the gas phase is considered adopting the κ-ε turbulence model developed by Benyahia et al. (2005) for disperse particulate flows. In dense fluidized beds, the solid stress tensor may be a function of kinetic/collisional and/or frictional contributions (Syamlal and Pannala, 2011). The kinetic/collisional solid stress is commonly calculated using the Kinetic Theory of Granular Flows (KTGF), while the frictional contribution is frequently obtained using rheological theories from soil mechanics (van Wachen and Almstedt, 2003). The KTGF, as reported in Agrawal et al. (2001), is used here to describe the solid stress tensor for volume fractions smaller than 0.42. Then, the stress tensors of the solid phases are estimated as functions of the so-called granular temperature. Here, the granular temperature is calculated by an algebraic equation (Syamlal et al., 1993; Syamlal and Pannala, 2011). For values greater than 0.42, the Schaeffer theory is used to describe the solid stress tensor (Syamlal et al, 1993). In the Eulerian approach, appears interaction terms between gas and solid phases in the momentum and internal energy conservation equations. For the momentum conservation equations, such term is called the gas-solid drag force and is taken into account here using the Gidaspow model (Gidaspow, 1994), while in the internal energy conservation equations, such terms are denominated as the interphase heat transfer model which are described here by the Ranz-Marshall correlation (Ranz and Marshall, 1952). It must be stressed that the energy transfer between different

solid phases is neglected (Syamlal and Pannala, 2011). The mathematical model is implemented using the MFIX (Multiphase Flow with Interphase eXchanges) open source code. (Syamlal et al., 1993).

3.2 Pyrolysis models

The pyrolysis model proposed by Miller and Bellan (1997), which is denominated here as MB model, is a generalization of the cellulose pyrolysis model by Bradbury et al. (1979), the so-called Broido-Shafizadeh model, and considers an initial reaction resulting in the conversion from an inactive state of the biomass components into an active one. The active components further decomposes into char, gas and tar through two competitive reactions. The last reaction considered in the model is the cracking of condensable tar into low-molecular-weight gases. However, the MB model does not address gas and tar compositions. The reactions scheme of the MB model is shown in Figure 1.

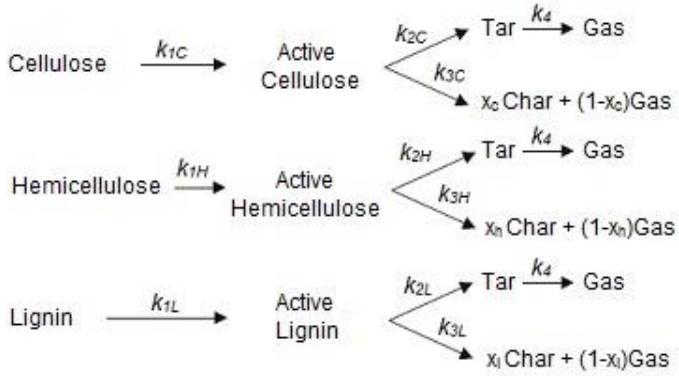

Figure 1. Biomass pyrolysis scheme of the MB model.

The model developed by Ranzi et al. (2008) is addressed in the present work as RAN model, which consists in a more complex set of reactions than the MB model. For example, the pyrolysis of a single component such as the hemicellulose contained in the biomass is considered to lead to two types of active hemicellulose. This model provides the compositions of gas and tar, and the corresponding stoichiometry. A simplified reactions scheme of the RAN model is shown in Figure 2.

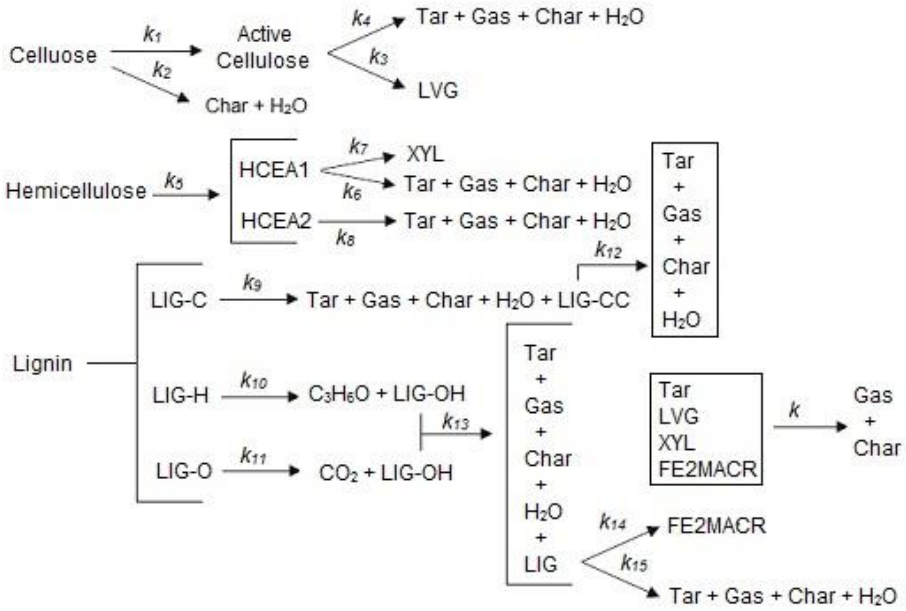

Figure 2. Biomass pyrolysis scheme of the RAN and BJR models.

Discussing the secondary pyrolysis of the tar compounds, the work of Blondeau and Jeanmart (2012) verifies that the RAN model is imprecise for high temperature situations, and thus proposes a modification to the model. Since some of the pathways in the RAN model are the same as those of the MB model, the modification proposed by Blondeau and Jeanmart (2012) consists in changing some of the RAN's kinetic constants by the respective ones given

in MB, resulting in the denominated here as BJR model. The reactions scheme of the BJR model is also given by the scheme presented in Figure 2.

Anca-Couce et al. (2014) proposes a modification of the RAN model that includes the secondary char formation reactions and the catalytic effect of alkali metals on sugar formation. The ANC model ignores the activation reactions, and includes the parameters x_{C1} for cellulose, x_{H1} and x_{H2} for hemicellulose, and x_{L1} and x_{L2} for lignin. Those parameters are used to change the products of some reactions of the RAN model. In general, the x parameters are functions of the gas retention time inside the particle, as well as the gas partial pressure and temperature, and the presence of minerals. In their simulations, however, Anca-Couce et al. (2014) simply adopt constant values for the x parameters. It is remarked that this simplification requires that these parameters be calibrated for each different situation.

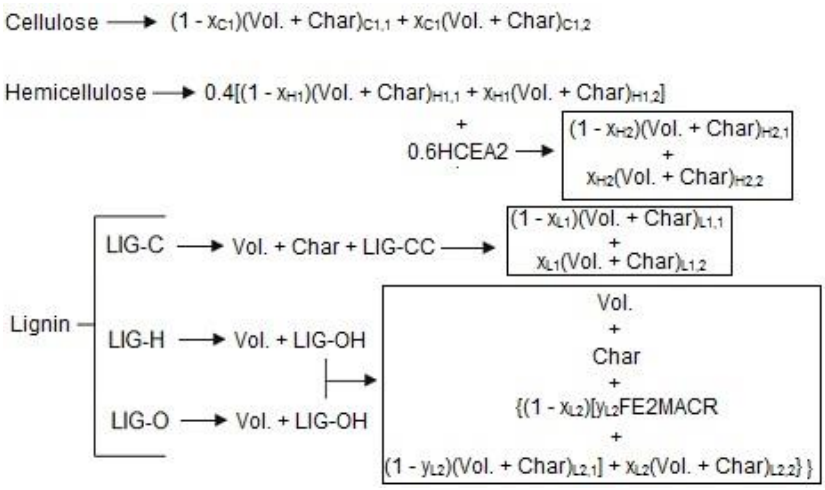

Figure 3. Biomass pyrolysis scheme of the ANC model.

In the pyrolysis scheme proposed by Gronli and Melaaen (2000), wood degrades into gas, tar and char by three parallel reactions. Tar then cracks into inert tar and gas as proposed by Boroson et al. (1989), giving the GMB model. The PAR model consists in the pyrolysis scheme proposed by Park et al. (2010). The PAR model assumes the thermal decomposition of wood in an intermediate solid, and its subsequent decomposition in char. Additionally, the authors also considered the presence of a tar re-polymerization reaction competing with the tar cracking reaction, yielding gas. Finally, SHA model, proposed by Sharma et al. (2014), is very similar to the GMB model, but the secondary tar reacts heterogeneously and leads to the formation of char by re-polymerization reactions at higher temperatures, as stated by Di Blasi (1993). It must be stressed that the GMB, PAR and SHA models consider a generic-type wood. Consequently, the models do not use the composition of the actual wood to predict the products of the pyrolysis. The pyrolysis reactions schemes of the GMB, PAR and SHA models are presented in Figure 4.

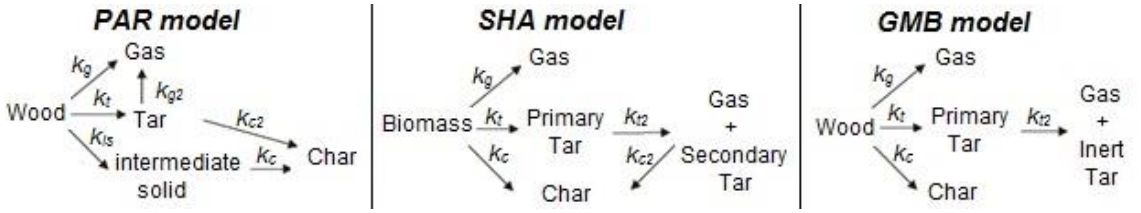

Figure 4. Biomass pyrolysis scheme of the PAR, SHA and GMB models.

The kinetic parameters used in the parallel reactions models denominated here as MB, RAN, BJR and ANC models are available, respectively, in the reference works of Miller and Bellan (1997), Ranzi et al. (2008), Blondeau and Jeanmart (2012) and Anca-Couce et al. (2014). For the kinetic parameters used in the competitive models, the parameters used in the GMB can be found in Gronli and Melaaen (2000) and Boroson et al. (1989), while the parameters used in PAR and SHA models can be found, respectively, in Park et al. (2010) and Sharma et al. (2014).

4. COMPUTATIONAL SETUP

The gasifier geometry adopted in the present work, also studied in Xue et al. (2012), is shown in Fig. 5. The reactor is 0.34 m in height and 0.038 m in diameter. The wood used in the simulations is red oak composed of 41% of cellulose, 32% of hemicellulose, 27% of lignin, and is free of moisture and ash. The red oak enters the reactor at a mass flow rate

of 0.1 kg/h and at a temperature of 300 K, while nitrogen is continuously injected at the bottom of the reactor with a velocity of 0.36 m/s and at a temperature of 773 K. The bed is formed by particles of biomass, char and silica sand. The properties of gases, tar and solids and physical parameters are taken from Xiong et al. (2013) and are shown in Tab. 1. The pyrolysis products leave the equipment at the top of the reactor. It is noteworthy that the biomass and nitrogen inlet flows have to be rescaled from the experimental 3D domain values to 2D domain values. The biomass inflow has been set to yield the same ratio of volume fraction per bed volume as the experimental inflow. The nitrogen velocity is kept the same, to assure identical fluidization characteristics.

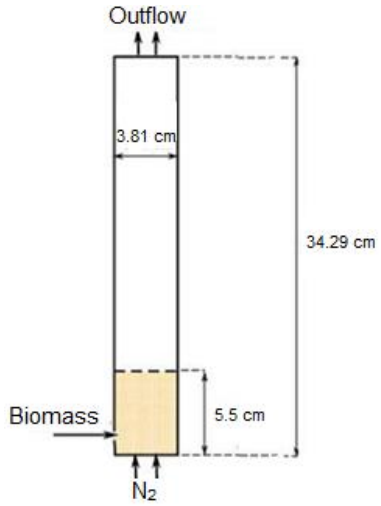

Figure 5. Two-dimensional Cartesian domain for the fluidized bed reactor.

Table 1. Physical properties of chemical species.

Specie	Density [g/cm^3]	Diameter [cm]	Specific heat [J/kg K]	Dynamic viscosity [kg/m·s]	Thermal conductivity [W/m·K]
Tar	-	-	2500	$3.00 \cdot 10^{-5}$	$2.58 \cdot 10^{-2}$
NCG	-	-	1100	$3.00 \cdot 10^{-5}$	$2.58 \cdot 10^{-2}$
N_2	-	-	1121	$3.58 \cdot 10^{-5}$	$2.58 \cdot 10^{-2}$
Biomass	0.4	0.04	2300	-	0.30
Silica sand	2.649	0.052	800	-	0.27

Numerical simulations are conducted considering the boundary conditions of no-slip wall for the momentum conservation equations, and zero mass flux for the species conservation equations. For the energy conservation equation, a constant wall temperature of 800 K is considered for the first 8 cm of height from the bottom, while adiabatic condition is assumed for the remaining of the reactor height.

A First Order Upwind (FOU) scheme is used for numerical discretization of the convective terms, while the first-order implicit Euler scheme is used for time discretization. A mesh with 10 x 90 volumes, identical to the one generated by Xiong et al. (2013), is used. In order to reduce the computational cost of the simulations, an adaptive time step is used. Here, as opposed to Xiong et al. (2013), constant apparent density and porosity of biomass particles are assumed.

5. RESULTS AND DISCUSSIONS

Table 2 shows the wood pyrolysis results from the present simulations and from the experiments reported in Xue et al. (2012). The indicated numerical results are taken as time averages at the outlet section of the reactor. Based on numerical experimentations conducted previously, the values of the x parameters to obtain the ANC model results presented in Table 2 are 0.0 for x_{C1}, 0.7 for x_{H1} and x_{H2}, and 0.0 for x_{L1} and x_{L2}.

For both flow regimes, it is observed in Table 2 that the MB model provides the best results for the pyrolysis of wood, leading to good estimates of the composition of the process products. The MB model also predicts correctly the temperature of the products leaving the reactor. The RAN model is known to overestimate the NCG yields, and to underestimate the tar yields at high temperature conditions (Blondeau and Jeanmart, 2012). The BJR model improves upon the RAN model with respect to the NCG and tar yields, but both models predict very similar results in relation to the amount of char produced and to the temperature of the gas phase leaving the reactor. Even the wood particle diameter being smaller than 1 mm, the mineral matter present in the biomass may affect the composition of the

pyrolysis products. The presence of mineral matter is not accounted for in the RAN and BJR models, but is taken into consideration through the *x* parameters used in the ANC model.

It is verified in Table 2 that the competitive reactions pyrolysis models GMB, PAR, and SHA predict similar temperatures for the gas phase leaving the reactor. However, the predicted amounts of gas, tar, and char differ remarkably. The GMB model significantly overestimates the amount of char yielded, resulting in the reduced production of NCG and tar. Both the PAR and SHA models predict similar amounts of char and gas phase exit temperatures. Nevertheless, while the PAR model overestimates the amount of NCG and underestimates the amount of tar, the SHA model has the opposite behavior.

Table 2. Comparison of products yields predicted by different pyrolysis kinetic schemes with experimental measurements.

	Flow Regime	NCG [%]	Tar [%]	Char [%]	T_{outlet} [K]
Experimental		19.2 – 21.8	70.3 – 73.1	11.5 – 14.5	773.0
MB	Laminar	19.3	64.5	16.2	774.0
	Turbulent	19.5	64.5	16.0	773.3
RAN	Laminar	31.6	60.4	8.0	751.4
	Turbulent	31.7	60.1	8.2	751.2
BJR	Laminar	25.9	64.3	9.8	754.9
	Turbulent	26.3	65.3	8.4	754.5
GMB	Laminar	13.0	54.0	33.0	771.0
	Turbulent	13.6	56.3	30.1	770.3
PAR	Laminar	25.9	65.8	8.3	773.7
	Turbulent	26.2	65.5	8.3	773.6
SHA	Laminar	15.1	77.4	7.5	774.4
	Turbulent	15.2	77.7	7.1	774.2
ANC	Laminar	24.1	58.0	17.9	755.7
	Turbulent	24.2	58.3	17.5	755.2

Results for simulations considering both the laminar and turbulent flow regimes are also reported in Tab. 2. For both flow regimes, no significant difference is observed. The highest relative difference was found for the char yield obtained with the GMB model, with a value of 8.7%. Nevertheless, for turbulent flow results, less unreacted biomass (not shown in Tab.2) is observed in the exiting streams, due to increased transport characteristics typical of turbulent flows, which helps to reduce the solids quantities yielded. Turbulence also has a small effect on tar cracking and tar generation reactions.

Figure 6 shows the temperature profiles along the reactor height for both flow regimes. It can be seen in Fig. 6 that for all kinetic models used, turbulence causes a slight decrease in temperature. A possible cause for that is the fact that turbulence enhances heat transfer among phases. Since most of the steps in each reaction model are endothermic, this decrease of temperature is present in the results obtained with all kinetic pyrolysis models studied here.

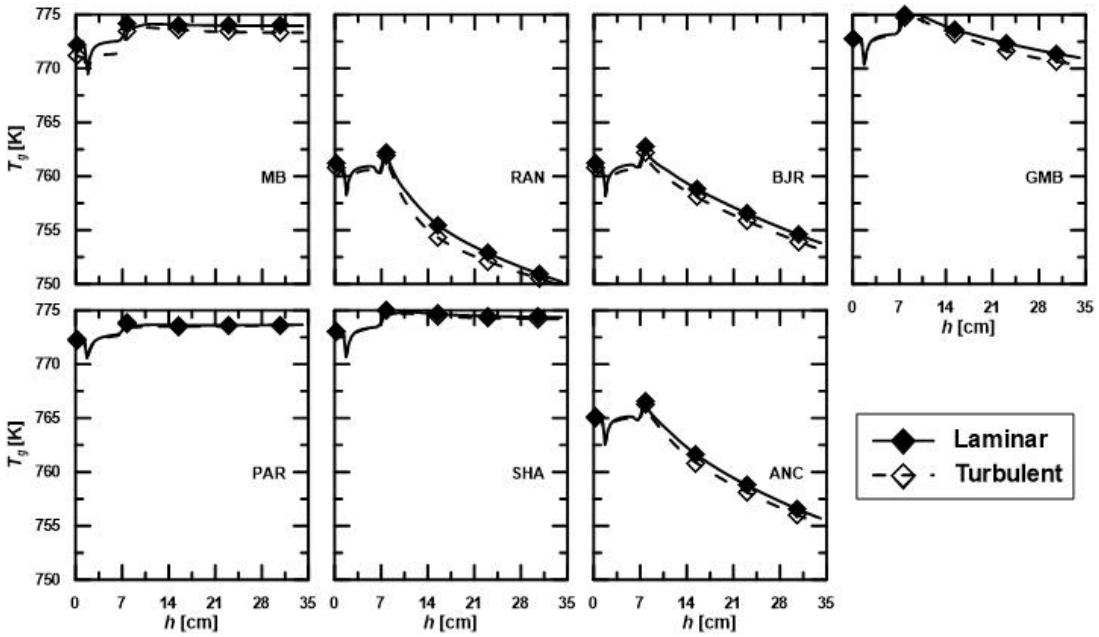

Figure 6. Temperature profiles along the reactor height for laminar and turbulent regimes.

Figure 7 shows the reaction rates for the formation of NCG and char and the net reaction rates of tar, for all the kinetic models studied. It can be seen in Fig. 7 that for the RAN, BJR, PAR and ANC models there is no significant deviance of the reaction rates between flow regimes. The SHA and GMB models provide similar behaviors, with turbulence inducing higher reaction rates. For the MB model, the laminar regime assumption delays the reaction rates. This can be deduced if observing that the curves for both flow regimes are similar in shape but, for the laminar regime assumption, the peak is at higher heights of the reactor, suggesting that there is a delay when it is assumed a laminar regime.

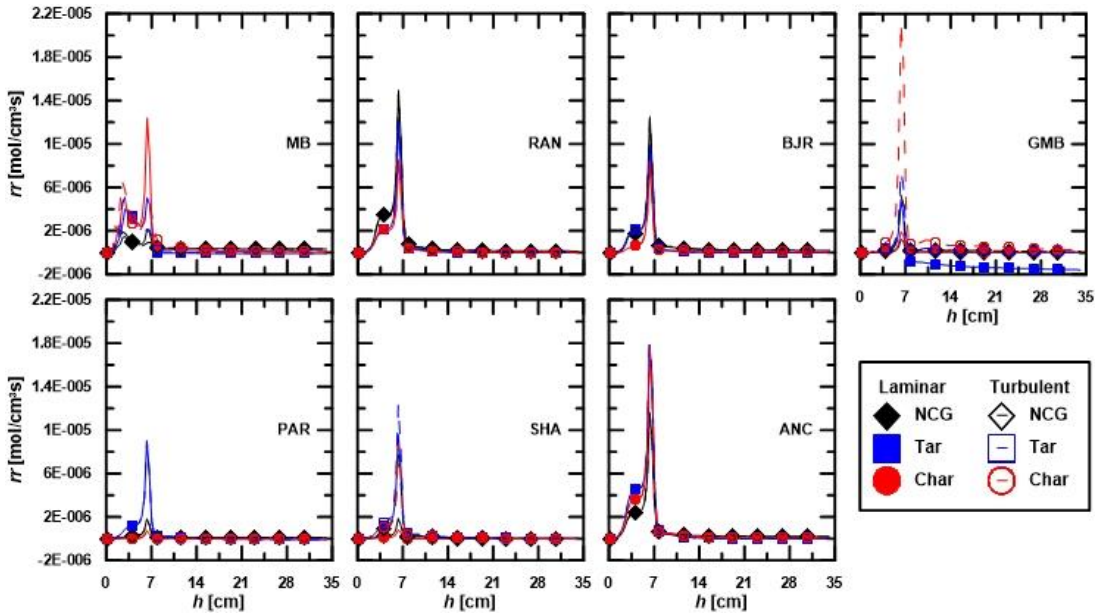

Figure 7. Reaction rates profiles along the reactor height for laminar and turbulent regimes.

6. CONCLUSIONS

The results revealed that the products distribution of wood pyrolysis is not significantly affected by whether one assumes the flow as laminar or turbulent. The results also revealed that when a turbulence model is used, the overall temperature inside the reactor decreases, except when the PAR and SHA kinetic models are used to describe the pyrolysis reactions. At last, it is shown that some kinetic models provide different behaviors for reaction rates profiles

depending on whether one assumes laminar or turbulent regime. While for the RAN, BJR, PAR and ANC models the reaction rates profiles is the same for both flow regimes, for the GMB and SHA models the reaction rates achieve higher values when assuming turbulent flow. At last, for the MB model the laminar regime assumption delays the reaction rates. However, in general, the presented results show only a small difference between predictions made using the κ-ε turbulence model and without consider any turbulence model. Then, since the use of κ-ε turbulence model implies the solution of two additionally transport equations, this work recommends not using the turbulence model to simulate fluidized bubbling bed reactors. Additionally, is also shown that de MB model provides the best prediction results for the wood pyrolysis process studied in the present work.

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